CHAPTER IX

BIOLOGICAL AND MISCELLANEOUS MONITORING STUDIES

INTRODUCTION

The previous chapters of this report reviewed the major historical environmental monitoring programs conducted routinely around the Rocky Flats Plant (RFP). Technically, the vegetation monitoring program could have been included in this chapter, as a biological medium. However, because vegetation was routinely monitored at the RFP during the time period of interest for this study, those data are addressed individually in Chapter V.

A variety of shorter term special studies involving biological samples (or biota) have been designed and conducted to understand transport pathways and distribution of materials released from the RFP. We have not attempted to collect or review all of these studies for this report. For example, a number of studies conducted by Colorado State University (Little 1976; Little et al. 1980; McDowell and Whicker 1978; Webb 1992) were designed to better understand the biological and physical transport mechanisms that redistribute soil containing plutonium (e.g., leaching, animal burrowing, resuspension, and runoff). However, for the purposes of the historical public exposures studies, the profile of plutonium concentration in soil at a point in time is of interest, but the mechanisms leading to that profile are not of practical interest. Relevant soil studies are reviewed in Chapter VIII.

A few studies are reviewed in relative detail in this chapter, either because members of the public and/or the Health Advisory Panel initiated that interest (e.g., study of actinides in cattle) or because *Radiological Assessment Corporation* (*RAC*) researchers felt that these studies would lend support to important conclusions drawn from other historical monitoring data (e.g., lichen study). Some other biological monitoring studies are only briefly mentioned for completeness.

REVIEW OF SPECIFIC STUDIES

Plutonium Concentrations in Lichens around Rocky Flats

Although lichens are a type of vegetation, this short-term research project is reviewed here rather than in the vegetation chapter (Chapter V), which addresses routine monitoring of vascular plants such as grasses. Unlike vascular plants, lichens obtain most of their nutrients from the atmosphere rather than their supporting substrate (typically soil, rock, or bark). The leafy morphology and relatively large surface area allow lichens to trap small airborne particles (visible with scanning electron microscopy) rather than requiring the elemental material to become soluble and be actively transported across cell membranes. Because lichens grow slowly, live long, and do not shed leaves or other parts, they are able to concentrate the substances they accumulate and, thus, provide a long-term reflection of environmental conditions (Rope and Pearson 1990).

The primary objectives of the lichen study conducted by Colorado State University were (a) to evaluate the concentrations of plutonium in lichens around the RFP as a function of distance and direction from the initial contamination site and (b) to correlate the lichen ^{239,240}Pu concentrations with available surface soil concentrations and provide a preliminary indication of the potential for lichen biomonitoring of the surface soil resuspension pathway (Thomas and

Ibrahim 1995). For the historical public exposures study, we are interested in examining the concentrations of plutonium in lichens around Rocky Flats because they may relate to the integrated plutonium concentrations in air at those locations. This provides another opportunity to examine spatial trends of plutonium released from the RFP. As compared to soil, lichens may reflect exposure to smaller particle sizes, whereas soil received deposition of all suspended particle sizes.

It is also advantageous for our study that the lichens are quite old. The lichens collected and

> **Lichens may reflect an integrated exposure to airborne plutonium during the time period of interest when measurements of plutonium in air are not available.**

analyzed in this study (74 samples in all) were aged and found to be 30–50 years old. Sample collections were made in 1991, so the specimens were in the environment as early as around 1940, 12 years before Rocky Flats operations began in 1952. Even the youngest lichen specimens were very likely to have been growing during the primary releases of plutonium from the barrel storage area (903 Area) in the late 1960s. There was no monitoring of plutonium in air near Rocky Flats during the late 1960s (see Chapter III). Thus, these lichen specimens provide an indication of plutonium distribution during an important time period when no

plutonium air monitoring data are available. Any spatial distribution of contamination found in the lichens should reflect the integrated spatial distribution of RFP releases, with the most important releases dominating the distribution pattern.

Lichen samples were collected from rock substrate within the buffer zone in directions ranging from roughly 40 degrees (approximately northeast) to 120 degrees (approximately east-southeast) and distances ranging from 200 m east of the 903 Pad to 3.0 km northeast near Indiana Street (Figure IX-1). Background lichen samples were obtained from an area northwest of Fort Collins, Colorado, over 70 km from the RFP. This distance is well beyond where Rocky Flats plutonium can be distinguished from fallout plutonium, even using sensitive analytical techniques (see Chapter VIII). Samples were ultrasound washed before analysis although split sample analysis of six samples showed no significant effect of washing on resulting plutonium concentrations. A tracer spike of ^{242}Pu was added to each sample to determine chemical yield. More details on the analytical methods can be found in Thomas and Ibrahim (1995).

The researchers concluded that there was a decrease in plutonium content of lichens with increasing distance from the RFP. They also reported a directional component (higher concentrations to the east) that supported conclusions of previous studies that windborne transport was the primary means of dispersion. In lichen sample region number 1 (adjacent to the 903 Pad, see Figure IX-1), the median concentration of plutonium in six lichen samples was 915 Bq kg^{-1} (25 pCi g^{-1}), with a range of 150 to 1280 Bq kg^{-1} (4.0 to 35 pCi g^{-1}). The median concentration in six background lichen samples was 1.30 Bq kg^{-1} (0.035 pCi g^{-1}) with a range of 1.18 to 1.80 Bq kg^{-1} (0.031–0.049 pCi g^{-1}). In three of the four sample regions bordered by Indiana Street on the east (numbers 13, 12 and 8, see Figure IX-1), the concentrations of plutonium in lichens were higher than in the background lichen samples. Concentrations were not statistically higher than background samples in sample region number 5, but only two samples were collected there.
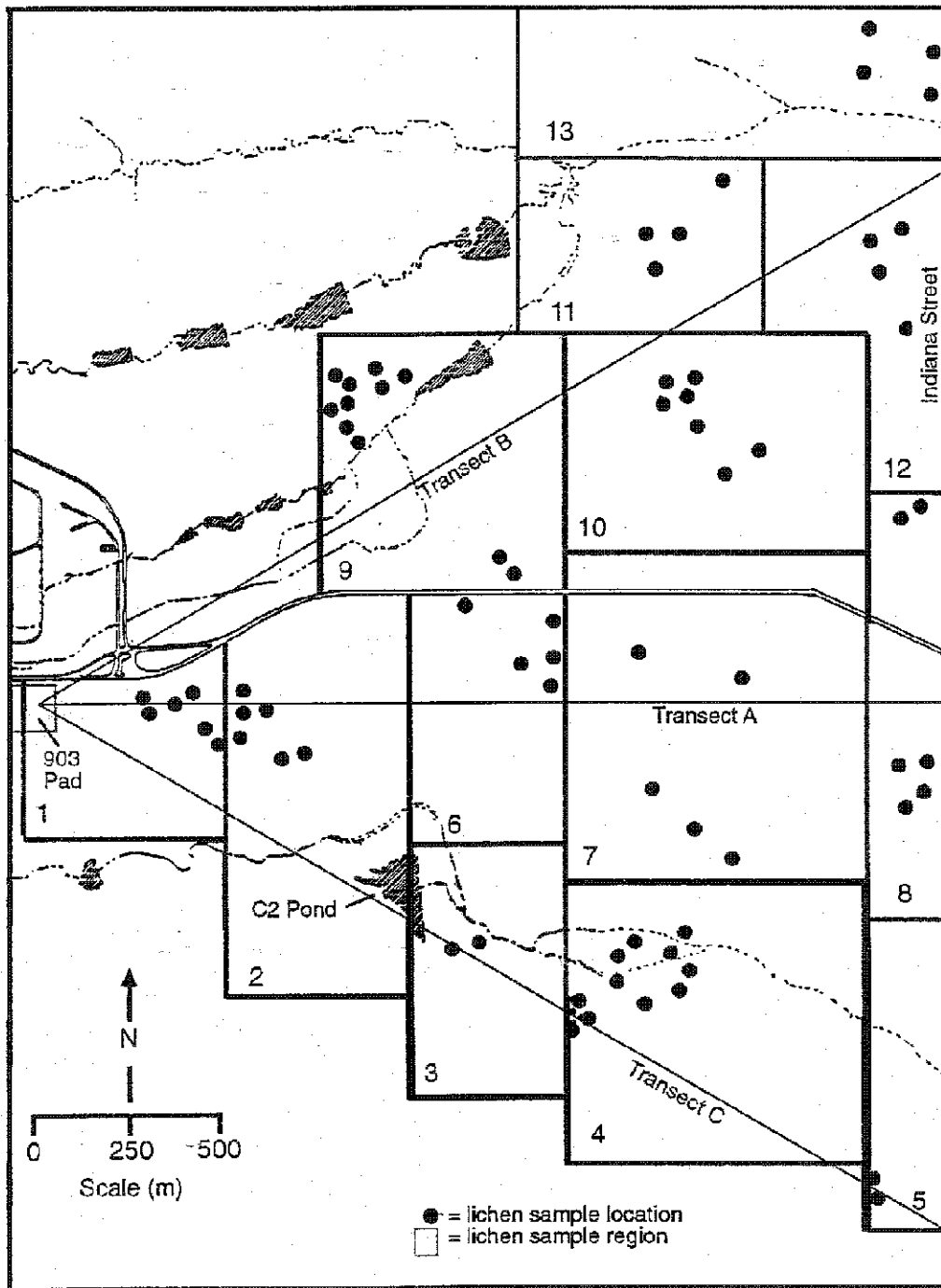
Figure IX-1. Map of lichen sample locations and regions east of the Rocky Flats Plant site (Thomas 1993).

Concentrations of plutonium in lichens were correlated with concentrations in soil from the same region (soil data from Litaor 1993, correlation coefficient, r = 0.77, significant at a probability of <0.001). However, the lichen-to-soil concentration ratio was highly variable. The concentration ratio tended to be lower near the 903 Pad, where plutonium contamination levels in soil were high and the associated particle sizes larger than farther away.

The plutonium concentration results in Thomas (1993) were presented in tabular form, organized by lichen sample region (n = 13) or by soil sample plots (n = 17) defined by Litaor (1993). A distance trend graph was generated for our report to assess the decrease in the concentration of plutonium in an easterly direction (Figure IX-2). Our initial graph was developed from the median concentrations in soil and lichen reported in Table 2 of Thomas (1993) and distances estimated from Figure 2 of Thomas (1993). Subsequently, the original data file was obtained from R.S. Thomas that contained distances computed from field location coordinates. Those original data are plotted in Figure IX-2, but the distances to soil data plots are still estimated from Thomas (1993).

Background concentrations of 0.035 and 0.048 pCi g^{-1} were subtracted from the lichen and soil concentrations, respectively, before plotting in Figure IX-2. This subtraction was necessary to evaluate the distance trend for Rocky Flats plutonium only. Background subtraction made very little difference for the soil measurements. Subtraction of background plutonium concentrations for measurements in lichens was important for collections made at distances greater than 2 km from the 903 Area.

Power-function trendlines were added to the chart, to help illustrate the concentration/distance trends for lichen and soil, using EXCEL software (Microsoft 1995). We are most interested in the relative decrease in plutonium concentration between the security fence (about 200 m from the eastern edge of the 903 Pad) and the boundary at Indiana Street (about 2.4 km from the eastern edge of the 903 Pad). This interest is because we have a daily air monitoring record (alpha activity) from the eastern security fence during the 903 Area disturbances, and we would like to be able to scale that integrated air concentration to a public access location(s). Using the trendline equations, there is a 200-fold decrease in the plutonium concentration in soil (from 300 to 1.5 pCi g^{-1}) over a distance of 0.2 to 2.4 km. For lichen over the same distance, the concentration decreases 80-fold, from 17.5 to 0.21 pCi g^{-1}.

The trendline for lichen shows a less rapid decrease in concentration than soil (Figure IX-2). However, a statistical test was not performed to see if the slopes of these two trendlines are statistically significant. McDowell and Whicker (1978) suggests that weathering or biological processes have broken down the particles with which the plutonium is associated. Thus, a particle-size gradient may exist where larger particles stay closer to the initial contamination and smaller particles become resuspended. In this sense, the lichen could have monitored a smaller range of resuspended soil particles. Relatively large particles transported by saltation, surface creep, or runoff would be measured in soil but perhaps would not be as likely to be incorporated into the lichen.

Subsequent to this lichen study, Webb et al. published a spatial model of plutonium in soil near Rocky Flats (Webb et al. 1997). They present equations for the drop-off in plutonium concentration in surface soil with distance from the pad in three directions (60, 90, and 120 degrees). The exponent of their power function equations was –1.40, –2.42, and –2.26,

respectively, for these three directions. The exponent of –2.13 for soil shown in Figure IX-2 falls within this range.
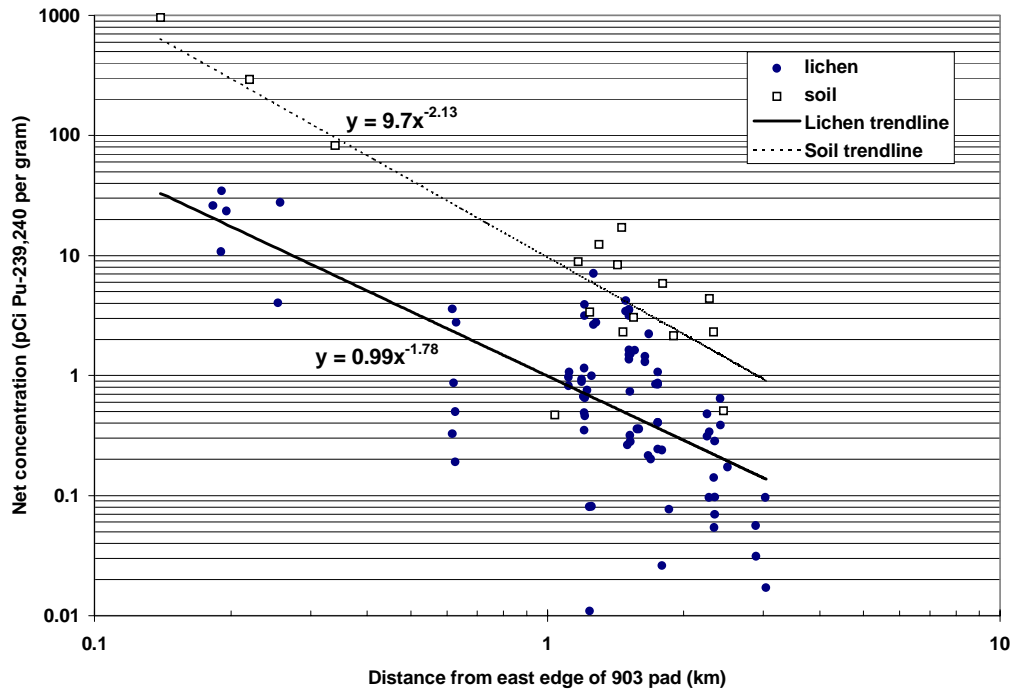

Figure IX-2. Trend in concentration of plutonium in lichen and soil with distance from the 903 Area on the RFP. Samples were collected in the buffer zone in directions that were generally northeast to southeast of the 903 Area. An estimate of fallout background was subtracted from the concentrations before plotting. Therefore, the points represent net concentrations from the releases of plutonium from Rocky Flats. The trendlines are power function fits to the data (Microsoft 1995).

　　　This distance trend for lichen and soil can be compared with plutonium in airborne particulates measured by the Health and Safety Laboratory (HASL) in the 1970s (see Chapter III). This air monitoring was conducted after the 903 asphalt pad had been established, so the airborne contamination was due to remaining contaminated soil areas. However, these areas were generally within several hundred meters of the pad (see Chapter VIII). Mechanical disturbances in the contaminated areas affected airborne contamination during this period as well. Figure IX-3 illustrates the spatial trend for these three media. The lichen and soil relative concentrations are estimated from the trendline equations in Figure IX-2.

　　　Langer (1991), in a summary of Rocky Flats resuspension studies in the early 1980s, gives the decrease in plutonium concentration in air with distance to the east during 1980–1985. The concentrations of plutonium in air at 1 m above the ground changed from 1.9 fCi m^{-3} in the 903 field (east of the pad and west of the security fence), to 0.48 fCi m^{-3} just east of the field, to 0.13 fCi m^{-3} at the vertical scaffold 100 m from the eastern edge of the field, and finally to 0.05 fCi m^{-3} at the cattle fence. Background fallout plutonium concentrations were around 0.005 fCi m^{-3} in the 1980s (see Chapter III). Thus, the relative concentration at the cattle fence (0.7 km east of

the 903 Area) to that near the security fence is (0.05–0.005)/(0.48–0.005), or 0.09, which is 4 times lower than the relative concentration from HASL measurements (0.38) in the 1970s (Figure IX-3). Disturbances like soil removal operations in the 1970s could have produced more resuspended plutonium compared to the 1980s.

The lichen data suggest that the time-integrated concentration of airborne particulates of Rocky Flats plutonium at Indiana Street, for the entire RFP operating history, might have been about 1% of that at the eastern security fence. Monitoring of air by the HASL after covering the pad would indicate an average concentration at Indiana Street that is about 3% of that measured at the eastern security fence, after global fallout background is subtracted. Temporal variability can be examined from the air monitoring data, which is not possible with the lichen measurements. The annual average ratio of the monthly plutonium concentration in air at Indiana Street ranges from 0.01 to 0.05 (1 to 5%) of that measured at the eastern security fence.

In summary, the lichen study data are useful for assessing risk from historical Rocky Flats releases in the following ways:

1. **Historical small-particle traps.** These lichens were present and trapping relatively small plutonium-bearing particles during most of the operating history of Rocky Flats (1951–1989). Even the youngest lichen samples were very likely to have been growing at the time of the highest releases from the 903 Area in the late 1960s. The only other integrating sample for which we have spatial data is soil.

2. **Distance trend (plutonium).** In combination with other environmental data, the trend of relative concentration of plutonium in lichen as a function of distance can be compared with the predicted trend in deposition of resuspended particles from our source term reconstruction and transport models. These data could be used either for model validation or calibration.

3. **Scaling time-integrated air concentration.** As a totally independent check on the dose calculation, 1–5% of the time-integrated activity concentration measured in air at the eastern security fence could be used for estimating cumulative exposure to a hypothetical person residing near the eastern boundary. A higher percentage would apply if a receptor were in the buffer zone during an accidental release.

4. **Risk comparison.** Concentrations in lichens from sampling regions that border Indiana Street are above concentrations measured in background lichen samples taken over 70 km away. (Thomas concluded they were statistically higher in three of four regions.) Because the background lichen samples were aged at 40–50 years old in 1991, they were exposed to the vast majority of weapons fallout plutonium, which was highest in the 1950s and early 1960s (see Chapter III). A median net concentration of 0.21 pCi g^{-1} at Indiana Street (Figure IX-2) is about 6 times higher than the concentration of fallout plutonium in background samples (0.035 pCi g^{-1}). By this crude yardstick, cumulative doses from Rocky Flats plutonium releases at the eastern boundary might have been about 6 times that from weapons fallout plutonium. This provides one perspective to the risk of plutonium releases from Rocky Flats and weapons fallout. Final risk comparisons will be incorporated into the risk assessment reports of Phase II of this study.
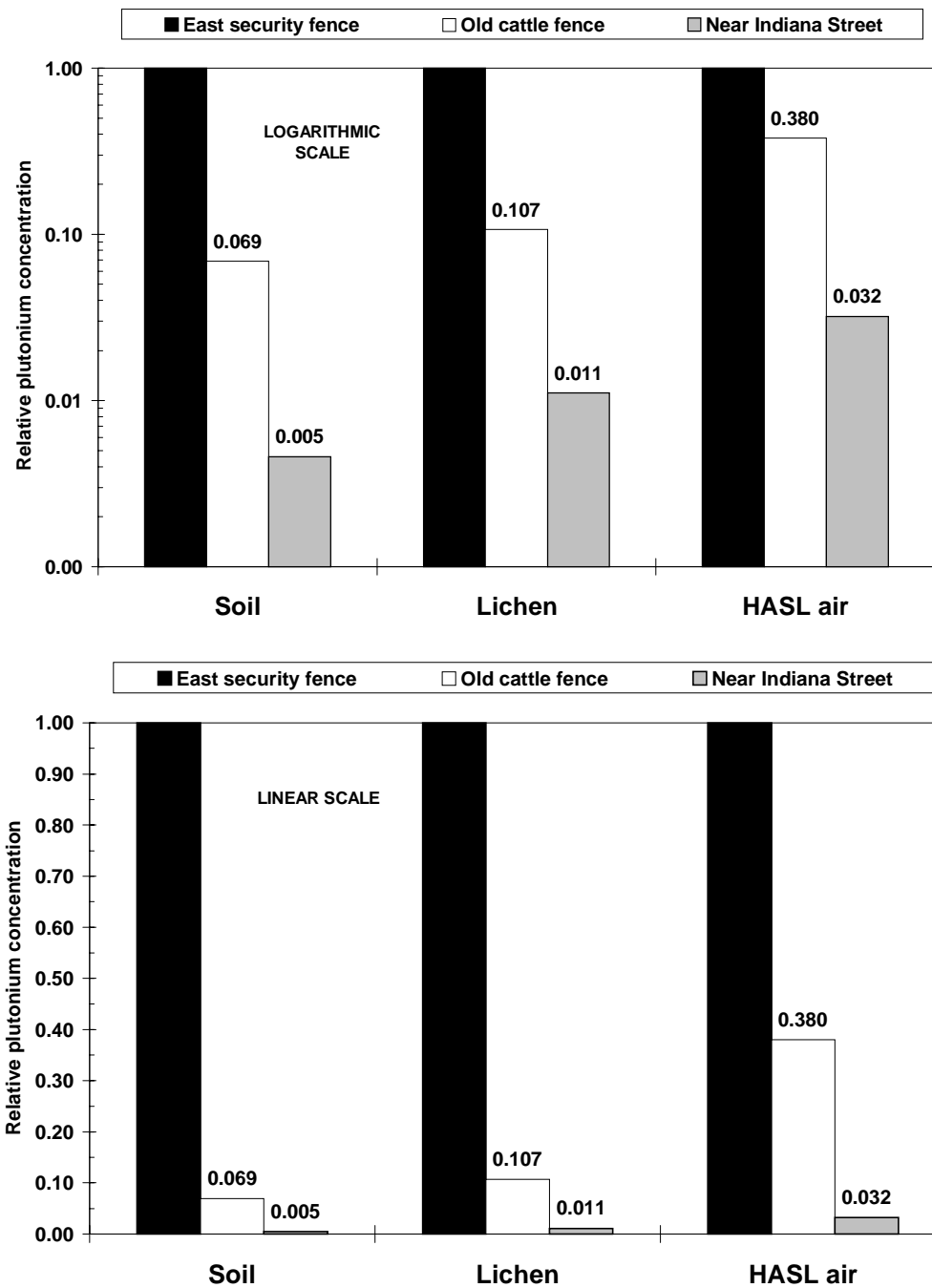
Figure IX-3. Distance trend in relative plutonium concentrations in three media. Concentrations at the two locations (0.5 km east near the old RFP boundary (cattle fence) and at the current boundary near Indiana Street) are presented as a fraction of the concentration measured in that medium near the security fence. Soil and lichen samples integrate plutonium releases over the entire history of RFP operations, whereas HASL air monitoring reflect conditions in the 1970s. An estimate of fallout background concentration was subtracted for all media, so the relative concentration represents the distance trend for Rocky Flats releases.

Pollen and Spores as Radionuclide Transport Vectors

Dr. Harvey Nichols, of the University of Colorado, under contract to the Energy Research and Development Administration, conducted a study of pollen and spores as radionuclide transport vectors. The purpose of the study was to determine (a) whether radionuclide particles in the size range of tenths of a micron (micrometer, abbreviated as Tm) are transported by larger particles, such as plant spores and pollen grains (size range several microns to 150 Tm), and (b) whether the low- and high-volume air samplers used at the RFP efficiently collected such larger particles. Large numbers of pollen and spores, released seasonally, travel long distances in air, attract other particles (by static charge), and carry the particles attached to elaborately sculptured (and sometimes sticky) pollen walls (Nichols and Nichols 1975; Nichols 1976).

Sampling began in July 1975. Static samplers for pollen were Vaseline-gel-coated plastic slides and gel-coated tapes attached to glass cylinders. These were placed at four main study areas in a northwest-southeast transect with respect to the Rocky Flats industrial area. One study area was the southeast perimeter fence near the 903 Pad. At a power pole southeast of the pad on the perimeter fence, the RFP provided an Anderson cascade impactor in a standard protective housing and a low-volume air sampler without a housing. The other three study areas were viewed as upwind and downwind controls. Pollen and air samplers were changed weekly for the first 8 weeks starting July 29, 1975, then biweekly as pollen release declined after September. Soil samples were collected at a variety of locations.

Samples of soil, fiberglass filters, and coated tapes were irradiated with neutrons in the research nuclear reactor at the Denver Federal Center. Polycarbonate film in close contact with the samples was etched and examined microscopically for fission tracks, then the position of fission tracks and pollen grains was examined. Star burst patterns resulted from particulates, whereas single tracks indicated dispersed radionuclides. A star at the same location as a pollen grain would implicate pollen as a transporter of the radionuclide. The authors state that cellulose nitrate film was used to establish that a particle was plutonium rather than uranium, but it is not explained how this is achieved. Cylindrical static pollen samplers were placed at 75-cm intervals from ground level up to 15 m on a meteorological tower at RFP to study the variation of pollen transport with height. The final report (Nichols 1976) states that very few fission tracks resulted from irradiation of these collectors. Therefore, the static samplers were abandoned in favor of reliance on the powered (air) samplers.

The progress report provided four initial conclusions: (1) RFP air samplers are not efficient for sampling large (50–100 μm) pollen particles, (2) large radionuclide particles have not been found attached to pollen; further time is needed to allow search for smaller particle attachment, (3) snow drift sites near Indiana Street have fissile particle concentrations about as high as the location near the 903 Area, (4) an elapsed time of 4 weeks is needed to allow autoirradiation of alpha-sensitive film placed on the soil surface at these locations to determine whether any of this activity is due to plutonium (Nichols and Nichols 1975).

The final report of this study (Nichols 1976) also stated that the association between pollen grains and fissile radioactive particles was negative. Thus, there was no indication that any significant number of radionuclide particles were transported by pollen grains or spores. The authors do conclude that the very abundant, light, and large pine pollen is not efficiently collected by low-volume ambient air samplers.

We make the following observations on the significance and conclusions of the pollen studies for assessing historical exposures of the public to Rocky Flats plutonium.

1. The hypothesis that plutonium particles might be transported long distances by large but buoyant pollen particles is an interesting one that was tested by these studies. However, the results of the studies did not show an association between pollen and radionuclides.

2. Small plutonium particles are transported long distances by themselves, without association with pollen or other organic substances. Those small particles are subject to dilution, dispersion, deposition, and blending into the worldwide fallout plutonium, so they cannot be easily distinguished from plutonium particles from other sources.

3. If plutonium particles had been associated with larger pollen particles, they would not be respirable, as reflected in the low collection efficiency of the samplers for the large pollen particles. If and when small particles disassociate from large particles, those small respirable particles would then be efficiently collected by ambient air samplers.

4. The fission track method used in the pollen studies does not identify the radionuclide causing the tracks. Dr. Nichols acknowledges in his reports that radiochemical methods are needed to be certain of the radioactive material being detected. He indicated in an interview (Nichols 1994) that he wanted to arrange for more in-depth analyses, but could not get support. The uranium mine 5 mi west of Rocky Flats is mentioned as the source of uranium, but all soils naturally contain uranium and thorium.

5. We believe that there is not enough consideration of the background and fallout to conclude that the fission tracks observed on samples from "accumulation areas," such as snow banks, are due to dispersion of Rocky Flats materials. We agree that nonuniform distribution of radionuclides occurs in the environment because of accumulating and scouring phenomena.

Measurement of Plutonium in Human Tissues from Autopsy

Dr. J.C. Cobb was a Professor of Preventative Medicine at the University of Colorado Medical School, a member of the Governor's Scientific Advisory Committee of Colorado, and a Member of the Air Pollution Control Commission of Colorado. Under a U.S. Environmental Protection Agency (EPA) contract, he investigated plutonium and americium in the tissues of people who have lived near the Rocky Flats plutonium plant near Denver. This study (Cobb et al. 1982) was conducted to determine whether the tissues of people who lived near to or downwind from the RFP contained more plutonium than the tissues of people who lived further away. Results were compared with two other studies of plutonium in human tissues. The sample size was over 400 individuals, after excluding those who might have been exposed to plutonium in the workplace or by proximity to atomic testing. Information was collected on the age, sex, smoking history, residence history, presence of disease, and plutonium levels in selected tissues from individuals autopsied in various Eastern Colorado hospitals. Various isotopes of plutonium were determined (mass numbers ^{236}Pu, ^{238}Pu, ^{239}Pu, and ^{240}Pu).

Summary findings from the study were that plutonium levels in lung and liver tissues were affected by age and smoking history more than by distance from the RFP. It appears from the graphs in the report that there were no individuals residing closer to the RFP than 10 km. Although the measured isotope ratio of ^{240}Pu to ^{239}Pu as a function of distance between 10 and 50 km indicated a small contribution from Rocky Flats, the total amount of plutonium in the samples was no different than in similar samples collected from other areas of the U.S. Age was the variable most closely related to the amount of plutonium in the liver (older people having more plutonium).

The Cobb study conclusions regarding the question of the RFP as a source of the plutonium were:

> . . . the results of this study are neither strongly positive nor strongly negative; one cannot conclude from these data that releases of plutonium from Rocky Flats have not contributed to human plutonium burdens, but the data do show that the total amount of plutonium in these human tissues is small and is not very different from that observed by other researchers who have studied people from other locations in the USA.

In a presentation to the Health Advisory Panel in February 1996, Dr. Otto Raabe commented that he had calculated dose from the tissue burdens of plutonium observed in the Cobb study and found them to be on the order of a few millirem. The amount of background radiation that an individual receives annually is several hundred millirem or more.

Colorado State University Urinalysis Study

This research was funded by the Colorado Department of Public Health and Environment and the Dow Chemical Company (Ibrahim et al. 1999). The work was intended to supplement other information to assess the risk to human health from manmade and naturally occurring radionuclides in the environment adjacent to the RFP. The objectives of this study were to estimate ^{239}Pu concentrations and excretion rates in urine of long-time residents living near the RFP compared to individuals living beyond any expected influence of the site. Because plutonium retained in the body from past exposure is excreted slowly in the urine, the concentration of plutonium in urine can reflect the amount of past exposure. This study also evaluated the significance of other variables including age, gender, smoking history, and lifestyle on plutonium levels in human urine.

Because individuals who have environmental levels of plutonium in their bodies excrete such small amounts in urine, very sensitive analytical techniques are required for detection. This study used fission track analysis (FTA), which is several order of magnitude more sensitive than alpha spectrometry, to measure very low levels of plutonium. FTA involves chemically isolating the plutonium from urine, bombarding with neutrons to produce a fission reaction from ^{239}Pu, and optically counting the fission tracks that result. One drawback of FTA is that plutonium tracks cannot be distinguished from the fissile ^{235}U. Thus any natural uranium contamination, if not properly separated from the sample, can interfere with (inflate) the plutonium results. Results presented in Ibrahim et al. (1999) document adequate separation of uranium and plutonium in the samples.

The study was conducted in two phases. An initial study involved collecting urine from April 1992 through December 1993 from 41 individuals not occupationally exposed to plutonium.[a] The Rocky Flats group included 10 adult females and 7 adult males living within 16 km of the plant, most of who reside to the east or southeast. The Rocky Flats participants were long-term residents; several were farmers or ranchers who had lived in the same locations for decades. The "background" group included 24 individuals who had always lived at least 16 km from the plant or any other similar contamination sources but were still in this area of Colorado. In the follow-up phase of the study, additional urine samples were subsequently obtained from another 23 nonsmoking adults between August and November 1995. There were 12 in the Rocky Flats group (three repeat individuals from Phase I) and 11 background group individuals. These background individuals were better matched with Rocky Flats participants in terms of gender and age when compared to the first phase of the study. All of these background participants in the follow-up phase lived in Larimer County (north of Denver), at least 60 km from the RFP.

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[a] None of the participants in either phase of this study had received occupational exposure to plutonium.

Measured levels of ^{239}Pu in urine from the Rocky Flats group were low and well within the range of reported "background" values, indicating small doses and low health risks. Only 53% of all measurements were at or above the detection limit. In the follow-up phase of the study, 10 of the 12 Rocky Flats participants had lived in the proximity of the RFP since 1964 and 8 of them since at least 1957. All had been local residents since 1968. The excretion rates of plutonium from the Rocky Flats group were higher than for the background group, but the difference was not statistically significant. There were two unusually high values, one in each of the Rocky Flats and background groups. The authors concluded that the fission track analysis technique might not be sufficiently accurate or precise to allow definitive comparisons between two groups of subjects with near-background levels of ^{239}Pu in urine.

Analysis of Tritium in Urine of Broomfield Residents

The Colorado Department of Health monitored tritium, a radioactive form of hydrogen, in the urine of Broomfield, Colorado, residents following an accidental release of tritium from the RFP in 1973. These data are presented in Chapter VI of this report along with a description of the release and other monitoring data.

Actinides in Cattle

A newsletter of the Environmental Information Network (EIN) included an article on actinides[b] in cattle grazing east of the RFP (Hurst and Elofson-Gardine 1993). The article was based on a study conducted by the EPA under a memorandum of understanding with the U.S. Energy Research and Development Administration (Smith and Black 1975). Following a discussion at the Health Advisory Panel meetings, *RAC* includes a review of this study here, even though the ingestion pathway risk is believed to be small compared to inhalation pathways.

Although the sample sizes were small (10 animals from Rocky Flats), the Smith and Black study contains important information about measured radioactive forms of plutonium, americium, strontium, hydrogen, and uranium in grazing cattle near the RFP. Cattle analyzed in the study were sacrificed in 1973. Five of the ten animals collected near Rocky Flats had grazed between mid-May and the end of October for the past 5 or 6 years on a 900-acre native grass pasture northeast of the plant and consumed water from Walnut Creek (Figure IX-4). The other five were raised at the same location but were only 5 months old, having been born in the spring of 1973.

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[b] Actinides – the heaviest elements, starting with actinium and continuing to the end of the periodic table. Some, such as uranium, are naturally occurring. Transuranic elements (for example, plutonium) are a subset of the actinide elements and include those with atomic numbers larger than uranium. Actinide elements are all radioactive.
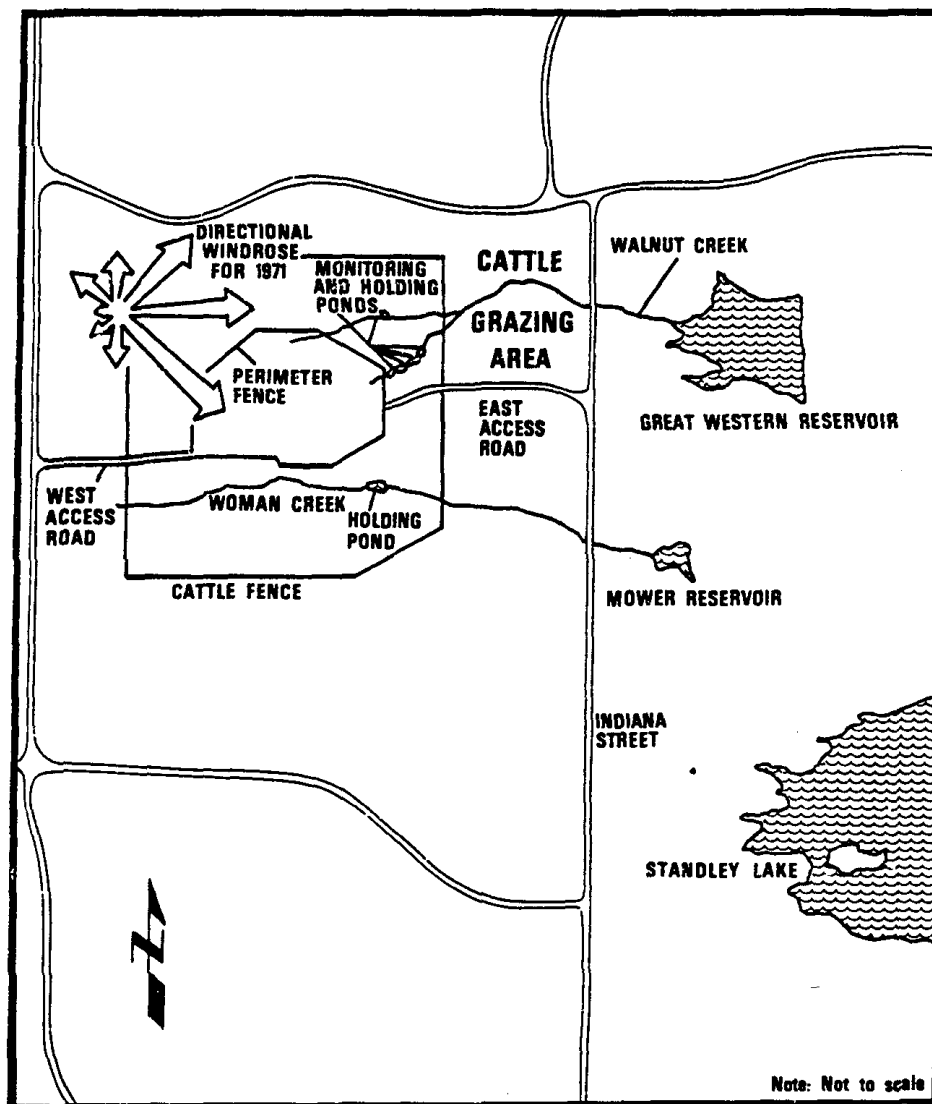
Figure IX-4. Location of pasture in which cattle grazed for 5–6 months per year between 1967 or 1968 through 1973, before analysis for actinide content of tissues (figure from Smith and Black 1975).

Concentrations of actinides in these Rocky Flats cattle were compared with those measured in cattle from Nevada, both near and distant to contaminated nuclear testing areas in that state (Figure IX-5). The cattle from Reno, Nevada, were collected from a slaughterhouse, and no other information is available for them. The Roller Coaster herd had access to range contaminated with plutonium during the 1963 Roller Coaster series of safety tests. The Nevada Test Site (NTS) herd grazed in an area containing the ground zeros of several atmospheric nuclear tests conducted in the early 1960s. The Searchlight herd grazed on a range that had been exposed only to worldwide fallout and the natural radioactivity of its soil. The animals from Nevada ranged over a wide area; therefore, exact exposure conditions were impossible to define.

Figure IX-5. Grazing locations of cattle collected from Nevada. NTS is the Nevada Test Site and TTR is the Tonapah Test Range (figure from Smith and Black 1975).

Smith and Black (1975) reported results of actinide concentrations in the tissues of the cattle in tabular and graphic form. As is the case with many studies of radioactivity in the environment, the concentrations measured were quite low, and statistical interpretations are problematic. The analytical laboratory reported their results in units of activity of each isotope per gram of ash. These original analytical data are reproduced in the appendices of the report. Smith and Black converted the concentrations to activity per gram of fresh weight based on the percent ash figures provided by the laboratory for each tissue. Plutonium results are plotted in Figure IX-6.
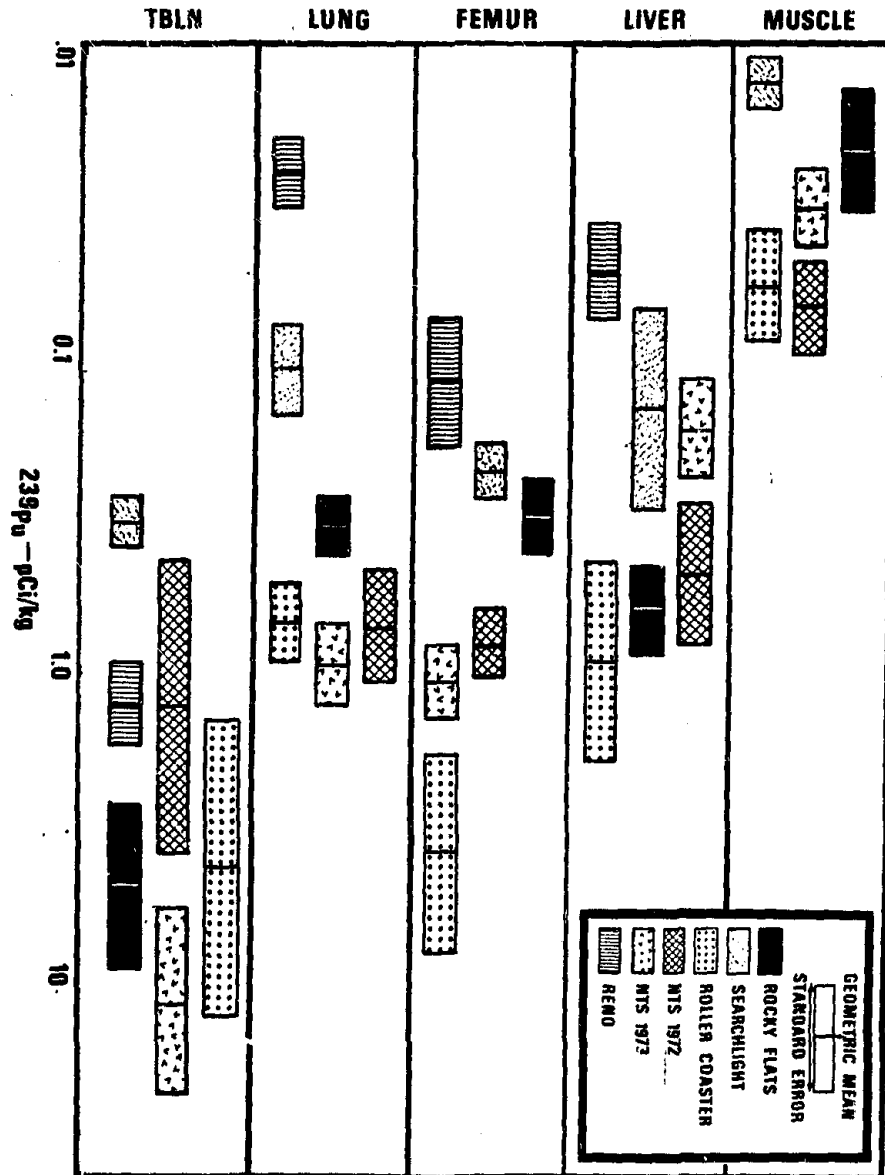

Figure IX-6. Results of analysis of plutonium in five tissues of cattle from Rocky Flats and five locations in Nevada. (figure from Smith and Black 1975).

It would be logical for the cattle obtained from the Searchlight and Reno locations to be relatively low and for the Roller Coaster and NTS herds to be relatively high (see locations in Figure IX-5). This appears to be the case. Concentrations of plutonium in Rocky Flats cattle fall somewhere in the middle of the groups sampled for all tissues (Figure IX-6). Sample size limitations, design flaws, and analytical limitations of this study are discussed or implied in their report. However, the authors conclude that "the data do suggest that the Rocky Flats cattle had a higher exposure to plutonium than the cattle from the Reno or Searchlight areas (background groups), and that this exposure was within the range of exposure of cattle on and around the NTS." The authors state that "the levels of both uranium and plutonium-239 found in the cattle are similar to those found in the general U.S. populations from fallout," and illustrate this fact for plutonium in Figure 4 of their report (reproduced as Figure IX-7 below).
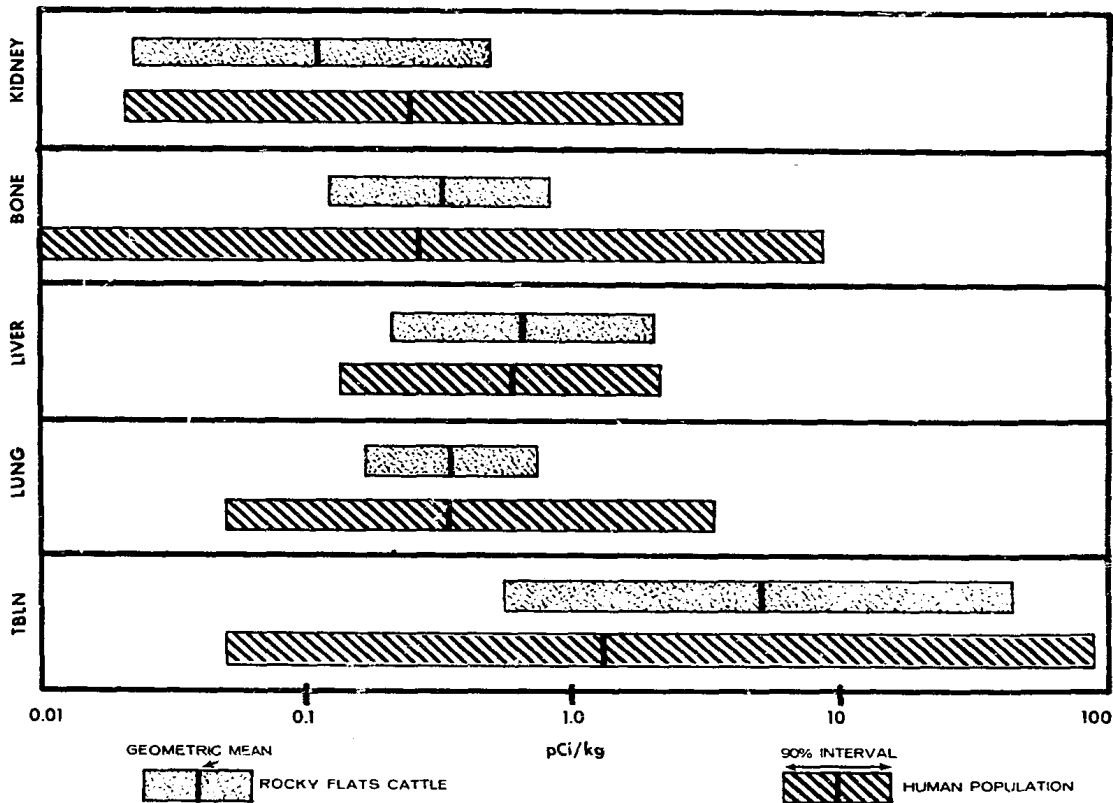

Figure IX-7. Comparison of plutonium concentrations in five tissues of Rocky Flats cattle with reported values in the human population of the U.S. (figure from Smith and Black 1975).

Uranium concentrations in cattle were within the range of previous studies, with the exception of tracheo-bronchial lymph nodes for the Rocky Flats cattle. The authors state that relatively high uranium concentrations would be expected there because of higher uranium content of soil found in the eastern slope of the Colorado Rockies.

A statement in the EIN article (Hurst and Elofsin-Gardine 1993) that americium concentrations were about "1/4 to 1/2 of the plutonium-239 concentrations in the same tissues" is excerpted verbatim from the Smith and Black report. The comparison is based on activity concentrations (i.e., picocurie of isotope per kg of tissue), not mass concentrations (i.e., picogram of isotope per kilogram of tissue). Water samples from Walnut Creek, which flowed through the Rocky Flats grazing area, showed plutonium concentrations about 8 times higher than americium. According to Phase I estimates of airborne releases from Rocky Flats, americium comprised 18% of the total long-lived alpha activity in effluents from plutonium areas, and ^{239,240}Pu contributed the other 82%. The ratio of ^{241}Am to ^{239,240}Pu activity in airborne effluents from plutonium areas was 0.22, or 22% (see Chapter III subsection titled "Apportioning Total Long-lived Alpha Count into Specific Radionuclides").

As a general rule, americium is slightly more biologically mobile than plutonium, but both are relatively immobile. For example, the root uptake of americium into plants has been stated to be about 10 times higher than for plutonium (Linsley et al. 1979; Garten 1978). However, soil-to-plant concentration ratios for uptake of plutonium are very low — ranging from 1×10^{-6} to 2.5×10^{-4} (ratio of concentration in undried vegetation to the concentration in dry soil), based on radioisotope experiments in plants grown in controlled environments (ATSDR 1990). Contamination of exterior plant surfaces might be expected to be relatively the same for plutonium and americium, and it can be 100 to 1000 times more than that resulting from root uptake (Romney et al. 1976). Following exposure to the same activity concentration in air, the activity concentration in cow's meat is predicted to be 2.4 times higher for americium than plutonium (CEC 1979). The ratio of americium to plutonium in the cattle tissue is not surprising considering the ratio released in Rocky Flats materials (~20%) and the higher biological mobility of americium.

Insight into the magnitude of this ratio in the general environment in the mid-1970s is gained from an article (Romney et al. 1976), which examined the ratio of ^{239,240}Pu to ^{241}Am in vegetation in 10 areas affected by fallout from the NTS and the Tonapah Test Range (TTR) (Figure IX-5). The nuclear tests of interest to these study areas occurred in 1954–1957 at the NTS and in 1963 at the TTR. The plutonium-to-americium ratio was "reasonably constant for vegetation samples collected from a given fallout area. This ratio, however, varied among separate test events primarily as the result of differences in the ingrowth of ^{241}Am within the aged source materials" (Romney et al. 1976). The mean ^{241}Am/^{239,240}Pu ratio in vegetation ranged from 0.08 to 0.25 at the NTS areas and from 0.06 to 0.08 at the TTR. The ^{241}Am/^{239,240}Pu ratio was 0.25–0.5 in the cattle tissues reported by Smith and Black. This somewhat higher ratio in the tissues might be due to the greater biological availability of americium relative to plutonium.

We did a dose calculation for a person consuming meat from cattle grazing east of Rocky Flats. The concentrations were taken from Smith and Black (1975) and ingestion dose factors used were those selected by ChemRisk (1994) for Phase I of this study. The committed effective dose equivalent from consuming 1/2 pound of meat per day containing the maximum measured concentrations of ^{239}Pu and ^{241}Am was computed to be 0.02–0.03 mrem following 1 year of ingestion. The calculation based on the geometric average measured concentrations was a factor of 10 less (0.002–0.003 mrem). Over 90% of the calculated dose was contributed by americium and less than 10% from plutonium. Some fraction of this total hypothetical dose would have been from globally dispersed fallout and not from Rocky Flats releases. There were no data reported for uranium in the muscle tissue, but relative concentrations for other tissues suggested that uranium would contribute a higher dose than plutonium or americium. This quick calculation supports the finding in Phase I that radiation doses from Rocky Flats-released materials from ingestion pathways would have been small relative to inhalation pathways.

Carbon Tetrachloride in Fish

Waterborne effluents were not historically monitored for any of the nonradioactive chemicals of concern for past public exposures (ChemRisk 1994). Although sampling of surface waters for carbon tetrachloride was not conducted, Zillich published a study of fish, macroinvertebrates, and water quality in surface water around the plant (Zillich 1974). Although the document states that no RFP materials are known to biomagnify in the food chain, the author also discusses bioconcentration of hydrocarbon solvents and seems to incorrectly assume that carbon tetrachloride and the other chlorinated hydrocarbons measured are insoluble in water and bioconcentrate in fish. (The log of the bioconcentration factor for carbon tetrachloride is 1.24 for trout and 1.48 for bluegill. A bioconcentration factor for bullheads has not been reported [Howard 1990]). Zillich compares his results to Food and Drug Administration standards for polychlorinated biphenyls in fish, standards that would not be applicable to carbon tetrachloride. Researchers tested several carp and a sunfish from Great Western Reservoir and detected no carbon tetrachloride in their tissues. Three white suckers taken from Walnut Creek were tested and carbon tetrachloride levels of less than 0.2 ppm were reported. Bullheads exposed for 2 weeks to water from South Walnut Creek and to water from further downstream where Indiana Avenue crosses Walnut Creek had carbon tetrachloride levels of 0.35 and 0.38 ppm, respectively. Unexposed fish had levels of 0.03 ppm (Zillich 1974). This is consistent with other reports of background levels, which range from 0.003 to 0.2 ppm but generally average about 0.02 ppm (Howard 1990). Zillich (1974) concluded that carbon tetrachloride concentrations in fish were insignificant. However, the levels of carbon tetrachloride in bullheads put into Walnut Creek water were about 10-fold greater than the levels in unexposed fish. In light of the fact that carbon tetrachloride has a low bioconcentration potential, these levels indicate that the fish were exposed to water with carbon tetrachloride concentrations as high as 10-fold greater than the concentrations in the waters in which the unexposed fish were placed.

Acknowledging that the [Zillich](#) (1974) study was done over a short time period with very few fish, these data can be used to estimate concentrations of carbon tetrachloride that may have been in the water in Walnut Creek at the time of the study. If we assume that an equilibrium was established between the fish and water in the creek, then the bioconcentration factor is equal to the ratio of the concentration in fish to the concentration in the water. If the carbon tetrachloride concentration in the fish was 0.38 mg L^{-1} and we apply the bioconcentration factors for trout and bluegill of 17 and 30, respectively, then the creek water concentration could be equal to 12 and 22 µg L^{-1}. If Walnut Creek provides from 2 to 4% of the total inflow into Great Western Reservoir, then the reservoir concentration might range from 0.20 to 0.8 µg L^{-1}. The drinking water standard (maximum contaminant level) set by the EPA is 5.0 µg L^{-1}. The ambient water quality criteria value for protection of human health from the water and from ingestion of fish and other organisms in the water is 4.0 µg L^{-1}.

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