APPENDIX H

EVALUATION OF BACKGROUND CONCENTRATIONS OF PLUTONIUM IN THE ENVIRONMENT AROUND THE ROCKY FLATS PLANT

This appendix provides further details about the information presented in <u>Chapter VIII</u> of this report. We describe sources of environmental plutonium around the Rocky Flats Plant (RFP), the isotopic composition of global fallout compared to Rocky Flats plutonium, temporal trends in global fallout, and measured background levels of plutonium in soils around the RFP and across the United States.

A variety of units have been used in the literature for expressing concentrations of plutonium in soils and other media. We have tried to avoid confusion by converting reported concentrations to SI units (becquerels per kilogram [Bq kg⁻¹], becquerels per square meter (Bq m⁻²], or becquerels per liter [Bq L⁻¹]) when comparing different sets of values. The units used in the original source document are also sometimes provided. In some cases, especially when we are mostly interested in the relative results from a single study, only the units from the original source document are provided. To convert values to different units, consult Table H-17 in the Annex to this appendix.

SOURCES OF BACKGROUND ENVIRONMENTAL PLUTONIUM

Global Fallout from Nuclear Weapons Testing

Atmospheric nuclear weapons testing is the largest source of plutonium in the environment (Harley 1979). Radionuclides formed in nuclear weapons tests are summarized in Holleman et al. (1987). Weapons-grade plutonium is composed primarily (weight-basis) of ²³⁹Pu, but it also includes ²³⁸Pu, ²⁴⁰Pu, ²⁴¹Pu, and ²⁴²Pu. In nuclear explosions, both fission and fusion weapons release plutonium; these releases are derived from unused plutonium (that does not fission) and from neutron capture reactions, which create the majority of the higher mass isotopes—²⁴⁰Pu, ²⁴¹Pu, ²⁴²Pu, and ²⁴³Pu. The quantities of ²⁴²Pu and ²⁴³Pu produced are very small. Very large quantities of ²⁴¹Pu are produced. However, ²⁴¹Pu decays primarily by weak beta emissions, and its radiological impacts are much less significant than those of the primary alpha-emitting plutonium isotopes—²³⁸Pu, ²³⁹Pu, and ²⁴⁰Pu. Other heavy-element radionuclides are also released in nuclear weapons tests, including ²³⁷U, ²³⁹Np, and ²⁴¹Am. Uranium-237 and ²³⁹Np have relatively short half-lives of about 7 days and 2 days, respectively, and they would not persist in the environment. Americium-241 builds up in the environment as a result of ²⁴¹Pu decay; therefore, it is present in the environment in significant quantities relative to the primary alpha-emitting plutonium isotopes. This appendix concentrates on the primary alpha-emitting plutonium isotopes²³⁸Pu, ²³⁹Pu. and ²⁴⁰Pu.

Eisenbud (1987) summarizes the history of nuclear weapons testing. Atmospheric testing of nuclear weapons started in New Mexico in July 1945. The majority of tests were performed by the U.S., former Soviet Union, United Kingdom, France, and China. Most of the atmospheric tests were performed in the 1950s and the early 1960s, before the signing of an atmospheric nuclear weapons test ban agreement in 1963 by the U.S., the former Soviet Union, and the United

Kingdom. The announced atmospheric tests are summarized in Table H-1, with estimated yields given in units of megatons of TNT, which would produce an equivalent explosive yield.

<u>Holleman et al.</u> (1987) provides a summary of the atmospheric transport of fallout from nuclear weapons testing. Because nuclear explosions create extremely high temperatures, a fireball is formed after the explosion. The expanding fireball can rise many kilometers, carrying debris from the explosion with it, and it reaches greater altitude for higher yield weapons. At low yields, from about 10 to about 200 kilotons, all of the debris remains in the troposphere (lower atmosphere). At high yields (1 to 2 megatons), 90 to 99% of the debris reaches the stratosphere. Transport of the material is dependent on the height at which the fireball initially injects the debris.

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Country	Period	Number of tests	Estimated total yield (megatons)						
U.S.	1945–1962	193	139						
Former Soviet Union	1949-1962	142	358						
United Kingdom	1952-1953	21	17						
France	1960-1974	45	12						
China	1964–1980	22	21						
Total		423	547						
^a Adapted from Eisenb	<u>ud</u> (1987).								

Table H-1. Summary of AnnouncedAtmospheric Nuclear Weapons Tests^a

Particles formed in the nuclear explosion can be transported long distances by winds. Material reaching the stratosphere is transported around the globe. Deposition of material from the atmosphere occurs by dry or wet deposition. To a great extent, air masses of the northern and southern hemispheres remain separated, but limited exchange between hemispheres does occur.

Other geographic features are relevant to global fallout in the area around the RFP. Mountains alter wind currents, resulting in a downward mixing of higher altitude air, which may increase the ground-level air concentrations of fallout on the lee side of the mountains (the side that is sheltered from the wind). High mountain passes and the lee side of mountains generally receive more precipitation than surrounding areas, which may increase the wet deposition of global fallout. For meteorological reasons, material from the stratosphere is transferred into the troposphere primarily over the middle latitudes (about 40° to 50° latitude) (UNSCEAR 1993). Thus, the greatest amounts of fallout from large tests, which reach the stratosphere, are eventually distributed in the middle latitudes, with lesser amounts distributed toward the poles and the equator (UNSCEAR 1993). The RFP is in the middle latitudes, at a latitude of about 40° north. Distribution of fallout from lower yield tests is dependent on the location of the explosion.

The small particles of debris from atmospheric nuclear weapons tests may remain in the atmosphere for quite some time. For material that reaches the stratosphere, residence times are generally determined to be about 2 to 4 years. Thus, tests that inject debris into the stratosphere generally do not produce the highest ground-level fallout concentrations until about 2 years after the explosions.

Global Fallout from SNAP 9A Burnup

In April 1964, a Transit Navigational Satellite was launched from California. Part of the payload was an auxiliary power generator (called SNAP 9A), which contained 17 kCi $(6.3 \times 10^{14} \text{ Bq})^{238}$ Pu (Harley 1979). The rocket system failed, and the satellite reentered the atmosphere in the southern hemisphere, burning up upon reentry at about 50-km (30-mi) altitude. Essentially all of the plutonium activity was ²³⁸Pu. The first arrival in the northern hemisphere of ²³⁸Pu fallout from the satellite burnup was measured in early 1966 in Italy. This source of plutonium contributes a small amount to the background total plutonium in the RFP area (see Table H-2 for a general comparison).

Localized Sources

A number of sources of localized plutonium exist in the environment, including both releases from nuclear processing facilities and releases from other accidents (<u>Harley 1979</u>). Releases from localized sources are confined to the lower atmosphere and, thus, are not globally distributed. The Nevada Test Site (NTS), in southern Nevada, was used for test detonations of small nuclear weapons up through 1961; safety tests in which the high explosives in nuclear weapons were detonated (with plutonium in the tested device); Plowshare explosions (using nuclear explosions for peaceful purposes); and accidental venting of underground weapons tests. A considerable amount of unfissioned plutonium was distributed from these tests. Material from the NTS was distributed at least as far away as Salt Lake City, Utah (about 600 km [370 mi] from the NTS), and it may have contributed small amounts to the plutonium deposition in Colorado.

The chemical explosives in nuclear weapons exploded in two incidents, following crashes of U.S. military aircraft. The first occurred in Palomares, Spain, in 1966, and the second in Thule, Greenland, in 1968. Both resulted in local dispersion of plutonium from the weapons.

A number of the U.S. atmospheric nuclear weapons tests were performed in the Pacific Ocean, at Bikini and Enewetak Atolls. Because large quantities of plutonium were produced in these tests, there was significant plutonium deposited in the local area around the tests.

France, India, and the United Kingdom have also released significant quantities of plutonium to oceans, in effluents and as packaged waste for disposal (<u>Harley 1979</u>). Essentially all of this material has remained in the oceans.

Location	Weapons ^{239,240} Pu	Weapons ²³⁸ Pu	SNAP ²³⁸ Pu					
Northern hemisphere Southern hemisphere	253 ± 33 67 ± 14	$6.1 \pm 0.8 \\ 1.6 \pm 0.3$	3.1 ± 0.8 10.3 ± 2.1					
Total	320 ± 36	7.7 ± 0.9	13.4 ± 2.2					
^a Based on measurements of plutonium from numerous locations (<u>Harley 1979</u>).								

Table H-2. Estimated Plutonium Inventories (kCi) in Soils in 1970^a

Several U.S. Department of Energy (DOE) weapons plants in the U.S. process plutonium, and releases have occurred from some of them (<u>Harley 1979</u>). At the Mound facility in Ohio, a liquid release of about 10 Ci $(4 \times 10^{11} \text{ Bq})$ of ²³⁸Pu occurred in 1969 from a break in a waste

pipeline. At the Los Alamos National Laboratory, in New Mexico, about 2 Ci $(8 \times 10^{10} \text{ Bq})$ of plutonium has been released to canyon waste disposal sites. Releases of plutonium have also occurred from the Hanford Site in Washington (TSP 1994), the Savannah River Site in South Carolina (RAC 1999), and to a lesser extent from the Oak Ridge National Laboratory in Tennessee (ChemRisk 1993). All of these facilities have been the subject of separate dose reconstruction projects and resulted in more or less localized plutonium deposition.

Table H-2 summarizes the estimated quantities of plutonium in soils in 1970, based on measurements at a number of locations in the northern and southern hemispheres, that were due to weapons tests and the SNAP 9A burnup (<u>Harley 1979</u>). These values are presented to give a rough indication of global inventories of fallout plutonium. In the northern hemisphere, background plutonium concentrations in soils primarily result from ^{239,240}Pu in weapons fallout.

ISOTOPIC COMPOSITION OF GLOBAL FALLOUT AND ROCKY FLATS PLUTONIUM

The plutonium processed at the RFP is weapons-grade plutonium, consisting primarily of ²³⁹Pu. Plutonium from atmospheric nuclear weapons tests is weapons-grade plutonium that has undergone (partial) fission and neutron capture reactions in the nuclear explosion. Because of these reactions, the relative abundance of the various plutonium isotopes is altered in the exploded material.

<u>Krey and Krajewski</u> (1972) measured the isotopic plutonium composition of a soil sample thought to contain plutonium essentially only from RFP releases and a sample from New York thought to contain only global fallout plutonium. Isotopic ratios, relative to ²³⁹Pu, are compared in Table H-3.

Table H-3. Mass Isotopic Ratios, Relative to ²³⁹ Pu, for Soil Samples Contaminated by Rock	ky
Flats Plutonium or by Global Fallout Plutonium	

Sample	²³⁸ Pu/ ²³⁹ Pu	$^{240}Pu/^{239}Pu$	$^{241}Pu/^{239}Pu$	²⁴² Pu/ ²³⁹ Pu
RFP plutonium	$7.15 \pm 10^{-5} \pm 9\%$	$5.10 \pm 10^{-2} \ \pm 0.19\%$	$2.31 \pm 10^{-3} \ \pm 0.42\%$	$1.43 \pm 10^{-4} \pm 1.7\%$
Global fallout	$1.35 \pm 10^{-4} \pm 5\%$	$1.80 \pm 10^{-1} \pm 0.24\%$	$7.76 \pm 10^{-3} \ \pm 0.5\%$	$3.89 \pm 10^{-3} \pm 0.59\%$
^a Source: <u>Kr</u>	ey and Krajewski (1972).			

Other sources have reported slightly different isotopic ratios for global fallout plutonium. <u>Krey</u> (1976) reported measurements for two samples from New York, with an average ratio 240 Pu/²³⁹Pu of 0.163 ± 0.008. <u>Bennett</u> (1978) reported measured plutonium isotopic mass ratios for stratospheric air samples for 1959–1970. The average measured ratio for 240 Pu/²³⁹Pu was 0.18, for 241 Pu/²³⁹Pu during 1963–1967 was 0.0138, for 241 Pu/²³⁹Pu during other years was 0.0118, and for 242 Pu/²³⁹Pu was 0.0034. While there may be slight differences in isotopic ratios in samples of global fallout plutonium, the isotopic ratios for RFP plutonium are significantly different than those for global fallout material. These significant differences can and have been used to differentiate between RFP and global fallout plutonium and to determine which source dominates in a particular soil sample (see also Table H-10). The most frequently used ratio is 240 Pu/ 239 Pu because the higher abundance of 240 Pu compared to other isotopes results in smaller uncertainties.

TEMPORAL TRENDS IN GLOBAL FALLOUT

In using background concentrations of plutonium in the environment for comparisons with concentrations near the RFP, it can be important to recognize temporal trends (changes with time) in global fallout. The major temporal trend in fallout plutonium concentrations is because of the timing of the weapons tests, which were the fallout plutonium source. <u>Bennett</u> (1978) summarizes the estimated explosive yields of all atmospheric nuclear weapons tests (see Table H-4). The cumulative yield is plotted later in Figure H-2.

Period	Total explosive yield	Cumulative yield
1945–1951	0.75	0.75
1952–1954	60.52	61.27
1955-1956	30.79 81.20	92.06
1957-1958	01.39 122 /3	295.88
1962	217.40	513.28
1964–1970	21.23	534.51
1971–1974	6.46	540.97
1976–1978	4.16	545.13

Table H-4. Summary of Estimated Total ExplosiveYields (Megatons) from Atmospheric Nuclear Tests

We examine the temporal trends in fallout plutonium by reviewing modeling predictions performed by <u>Bennett</u> (1978). We do not rely on these predictions for explicit, quantitative uses; they are presented to give an appreciation of the general trends.

Bennett (1978) used this information about the timing of weapons testing, the locations of the detonations, and an atmospheric transport model to predict fallout concentrations of plutonium and americium in surface air in the middle latitudes of the northern hemisphere. Table H-5 shows the predicted air concentrations of ^{239,240}Pu, and Figure H-1 is a plot of these concentrations. The air concentrations of plutonium from global fallout vary considerably over time. It is important to consider this temporal trend of air concentrations. Because of the seasonal changes in mixing of air masses, there are also seasonal trends in fallout air concentration measurements, these seasonal trends should be considered. Because our major focus of this report is soil samples, the seasonal trends are not examined in more detail.



Figure H-1. Predicted surface air concentrations of ^{239,240}Pu (fCi m⁻³) from global fallout from nuclear weapons testing. Predictions are for the mid-latitudes in the northern hemisphere.

Year	Concentration	Year	Concentration	Year	Concentration
1950	0	1961	0.089	1972	0.032
1951	0.0001	1962	0.54	1973	0.021
1952	0.0022	1963	1.18	1974	0.028
1953	0.031	1964	0.58	1975	0.017
1954	0.097	1965	0.25	1976	0.0083
1955	0.16	1966	0.11	1977	0.044
1956	0.14	1967	0.054	1978	0.018
1957	0.19	1968	0.042	1979	0.0071
1958	0.25	1969	0.056	1980	0.0028
1959	0.33	1970	0.062		
1960	0.11	1971	0.066		

Table H-5. Predicted Surface Air Concentrations of ^{239,240}Pu (fCi m⁻³) in Mid-Latitudes of the Northern Hemisphere from Global Fallout from Nuclear Weapons Testing^a

Bennett (1978) also used the atmospheric transport model to predict deposition rates and cumulative deposition of plutonium and americium in the New York region. These predictions are shown in Table H-6. The predicted cumulative deposition of ^{239,240}Pu is plotted in Figure H-2, along with the cumulative yield of the weapons tests for comparison. The predicted cumulative deposition follows the same general shape as the cumulative yield after the lag time because the residence time of the material in the stratosphere is accounted for. The temporal trend in the cumulative deposition of fallout plutonium should be considered when comparing RFP-influenced soil sample results with background results. This trend can be especially important when comparing samples taken at different times. We note that the predicted cumulative deposition of plutonium from nuclear weapons fallout reaches about 90% of its predicted maximum value in 1968, and it reaches 95% of maximum in 1971 (this is relevant to the background soil samples discussed later in this appendix). Although these predicted depositions are in the middle latitudes.

Year	Deposition rate (mCi km ⁻² y ⁻¹)	Cumulative deposition (mCi km ⁻²)	Year	Deposition rate (mCi km ⁻² y ⁻¹)	Cumulative deposition (mCi km ⁻²)
1950	0	0	1968	0.021	2.00
1951	0.00006	0.00039	1969	0.047	2.04
1952	0.0012	0.0013	1970	0.031	2.07
1953	0.017	0.019	1971	0.029	2.10
1954	0.054	0.072	1972	0.023	2.13
1955	0.091	0.16	1973	0.017	2.14
1956	0.077	0.24	1974	0.018	2.16
1957	0.11	0.35	1975	0.012	2.17
1958	0.14	0.49	1976	0.0075	2.18
1959	0.19	0.67	1977	0.024	2.20
1960	0.061	0.73	1978	0.0098	2.21
1961	0.049	0.78	1979	0.0039	2.22
1962	0.30	1.08	1980	0.0016	2.22
1963	0.44	1.52	1981	0.00061	2.22
1964	0.26	1.78	1982	0.00022	2.22
1965	0.11	1.89	1983	0.00011	2.22
1966	0.046	1.93	1984	0.00006	2.22
1967	0.042	1.98			

Table H-6. Predicted Deposition Rate and Cumulative Deposition of ^{239,240} Pu in the New
York Area because of Global Fallout from Nuclear Weapons Testing ^a

^{*a*} Source: <u>Bennett</u> (1978).



Figure H-2. Predicted cumulative deposition of plutonium in the New York area because of global fallout from nuclear weapons testing. For comparison, the cumulative explosive yield of atmospheric weapons tests is also plotted.

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BACKGROUND PLUTONIUM IN SOIL NEAR THE ROCKY FLATS PLANT

This section describes results from studies around the RFP that represent background concentrations of plutonium in soil. The fire at the RFP in 1969 caused an increased interest in monitoring soil concentrations of plutonium around the plant. A number of soil monitoring studies around the plant were performed or begun in late 1969 and in the early 1970s. Studies were performed by the National Center for Atmospheric Research (NCAR) for the Colorado Committee for Environmental Information (CCEI), the Colorado Department of Health (CDH), the Health and Safety Laboratory (HASL) of the Atomic Energy Commission, and Colorado State University (CSU). In addition, a study by the RFP of background concentrations involves sampling at 50 locations.

The NCAR and CDH obtained surface samples (to 1-cm [0.4-in.] maximum depth) and reported results as mass concentrations, while HASL obtained samples to 10 and 20-cm (4 and 8-in.) depths and reported results as total deposition (per unit area). The shallow depths of the NCAR and CDH samples mean that not all of the plutonium in the soil column was sampled. It is not reasonable to convert the mass concentration results of NCAR and CDH to total deposition values; thus, the NCAR and CDH results cannot be directly compared with HASL results. The CSU study used both surface and deeper soil samples and developed models to describe the soil concentrations of plutonium as a function of depth.

National Center for Atmospheric Research Study

The first study was performed by NCAR for CCEI in late 1969 and early 1970. Results were reported first by CCEI (CCEI 1970), with additional results given in the later report by NCAR (Poet and Martell 1972). This study sampled soils at 35 locations around the RFP and in the Denver area and three locations on the eastern slope of the Rocky Mountains that were thought to contain plutonium only from nuclear weapons fallout. For this study, surface soil samples were taken to a depth of 1 cm (0.4 in.). The background sampling locations are shown in Figure H-3. Results from the background locations are provided in Table H-7. Analysis errors (standard deviations) are included to provide general perspective on the analytical precision. Results were given in units of disintegrations per minute per gram (dpm g^{-1}), and we have converted these to becquerels per kilogram (Bq kg⁻¹) in Table H-7.



Figure H-3. Background sample locations in Colorado established by the National Center for Atmospheric Research, Colorado Department of Health, and Colorado State University soil studies (after the 1969 fire).

CDH Sampling

Soil sampling around the RFP was also performed by the CDH. Results of monitoring for 1970–1977 are presented by CDH (<u>1977</u> and <u>1990</u>) and <u>Jones and Zhang</u> (1994). Samples were generally collected from 13 sectors near the RFP and up to nine remote sites in Colorado each year, although in some years not all the sites were sampled. The remote site locations are shown in Figure H-3. The CDH used its own method to obtain samples for 1975–1988. This method included taking 25 individual surface samples at each site and then compositing to form a single sample for analysis. The sampling procedures used for years before 1975 were not detailed. The sampling depth has changed over the years, with depth 0.16 cm (0.06 in.) used for 1970–1974, 0.32 cm (0.13 in.) for 1975–1981, 0.48 cm (0.19 in.) for 1986, and 0.64 cm (0.25 in.) for 1989 and 1991 (Jones and Zhang 1994). Results from the background locations for 1976 and 1977 are provided in Table H-8. Analysis errors (2σ) are also shown to provide general perspective on the analytical precision. Results for 1970–1991 are summarized in Table H-9, although no results for these background locations were available for 1974, 1979, 1981–1985, 1987, 1988, and 1990. Results were given in units of disintegrations per minute per gram, which we converted to becquerels per kilogram in Tables H-8 and H-9.

	Concentration of ^{239,240} Pu ^a						
	dpm	n g ⁻¹	Bq kg ⁻¹				
Location	Value	Std. dev. ^b	Value	Std. dev. ^b			
Loveland	0.047	0.013	0.78	0.22			
Loveland	0.056	0.025	0.93	0.42			
Loveland	0.045	0.008	0.75	0.13			
Loveland	0.026	0.006	0.43	0.1			
Loveland	0.043	0.005	0.72	0.08			
Brighton	0.093	0.009	1.6	0.15			
Cripple Creek	0.140	0.027	2.3	0.45			
Cripple Creek	0.052	0.012	0.87	0.20			
Cripple Creek	0.117	0.015	2.0	0.25			
Mean ^c	0.069		1.1				

Table H-7. Background (Fallout) Concentrations of Plutonium in Surface Soil (0–1 cm [0–0.4 in.]) Measured by the National Center for Atmospheric Research around the Rocky Flats Plant in 1969–1970

^a The source document (<u>Poet and Martell</u> 1972) gives results in units disintegrations per minute per gram (dpm g^{-1}).

^b Std. dev. = standard deviation.

^c The arithmetic mean has been calculated, in this present work, from the individual values.

Table H-8. Background (Fallout) Concentrations of 239,240 Pu in Surface SoilMeasured by the Colorado Department of Health in Colorado in 1976 and 1977(Bq kg⁻¹)^a

	1976		1977		
Location	Value	Counting error (2Φ)	Value	Counting error (2 Φ)	
Burlington	0.3	0.3	1.2	0.3	
Crooke	1.3 ^b	0.2	0.7	0.3	
Limon	0.7	0.3	0.7	0.3	
Livermore	0.3	0.3	< 0.3		
Loveland	с		0.3	0.3	
Penrose	1.5	0.7	0.7	0.3	
Springfield	0.3	0.3	0.3	0.3	
Walsenburg	с		0.7	0.7	

Values were reported in units disintegrations per minute per gram in the source document ($\underline{CDH 1977}$). Sampling depth for these years was 0.32 cm (0.13 in.).

^b Average of two samples.

^c No sample was taken at this location in 1976.

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Location	1970	1971	1972	1973	1975	1976	1977	1978	1980	1986	1989	1991
Burlington	1.5	1.8	1.2	0.8	b	0.3	1.2	< 0.7	0.7	< 0.3	1.7	0.5
Crooke	0.7	2.2	1.8	0.8	b	1.3	0.7	1.2	b	< 0.3	0.7	0.7
Huerfano Butte	b	b	b	b	b	b	b	b	b	b	b	2.3
Limon	2.2	1.0	1.2	1.0	b	0.7	0.7	< 0.7	0.7	<1.2	0.5	1.0
Livermore	0.7	< 0.7	1.2	0.7	b	0.3	< 0.3	0.7	b	< 0.7	< 0.3	1.3
Loveland	1.8	1.7	b	2.0	b	b	0.3	< 0.7	b	< 0.7	< 0.3	< 0.3
Penrose	1.8	1.3	1.8	1.0	b	1.5	0.7	2.2	b	< 0.7	0.3	2.5
Springfield	0.7	1.5	2.0	< 0.7	0.7	0.3	0.3	4.5	0.7	<1.5	< 0.2	1.3
Walsenburg	1.8	1.2	1.8	0.8	0.3	b	0.7	< 0.7	b	< 0.7	1.7	0.8

Table H-9. Background (Fallout) Concentrations of ^{239,240}Pu in Surface Soil Measured by the Colorado Department of Health in Colorado in 1970–1991 (Bq kg⁻¹)^a

^a Values were given in units of disintegrations per minute per gram in the source documents (<u>CDH 1977, CDH 1990</u>; <u>Jones and Zhang 1994</u>). Sample depths were 0.16 cm (0.06 in.) for 1970–1974, 0.32 cm (0.13 in.) for 1975–1981,

0.48 cm (0.19 in.) for 1986, and 0.64 cm (0.25 in.) for 1989 and 1991.

^b No sample result was available for this location for the indicated year.

Health and Safety Laboratory Studies

Studies of plutonium in soil around the RFP by the HASL of the U.S. Atomic Energy Commission were initiated in early 1970. These studies did not separately select background sampling locations as done in the studies described above. Instead, sample locations were chosen at increasing distances from the RFP, and calculation techniques were generally employed to estimate background concentrations. The first study by the HASL is reported by Krey and Hardy (1970). Samples were collected in February 1970 from 33 sites around the RFP, to distances of about 64 km (40 mi), and primarily in easterly directions from the site. Figure H-4 shows the numbered locations (1-33) except for some of those close to the plant. Samples were collected to a depth of 20 cm (8 in.). At some locations, depth profile information was obtained by collecting samples in incremental layers to a total depth of 20 cm (8 in.). Based on limited depth profile information, Krey and Hardy concluded that less than 1% of the total plutonium in soil was deeper than 13 cm (5.1 in.). Results from this study were expressed in units millicuries per square kilometer total deposited plutonium based on the assumption that the measured plutonium (to depth of 20 cm [8 in.]) was the total deposited plutonium. Because the studies described earlier in this section used shallow sample depths, their results cannot be reasonably compared to results of these HASL studies.

<u>Krey and Hardy</u> (1970) did not measure or calculate background plutonium concentrations in soil from their 1970 sampling. They report a background concentration of 1.5 mCi km⁻² (56 Bq m⁻²) based on a single measurement in 1965 in Derby, Colorado (Figure H-4).

Seed et al. (1971) performed additional analyses on the data of Krey and Hardy to estimate the background plutonium concentration. Seed et al. plotted the distribution of measured concentrations on log-probability paper. This plot indicated that the distribution appeared to be made up of two separate lognormal distributions: one that represented samples dominated by RFP material and one that represented samples dominated by worldwide fallout plutonium. The data were separated into these two subgroups and replotted. Straight lines (on log-probability plots) were fitted to the data to obtain statistics about the distributions. From the fitted lognormal distribution, we determined the background distribution to be represented by a median



Figure H-4. Locations of the Health and Safety Laboratory soil sampling around the Rocky Flats Plant. Locations 1–33 were used in the 1970 sampling (Krey and Hardy 1970; Seed et al. 1971) and the 1971 sampling (Krey and Krajewski 1972). Locations 34–43 were added in the 1972 sampling (Krey 1976). Only locations numbered higher than 22 are shown here. Other locations are close to the plant.

concentration of 2.3 mCi km⁻² (85 Bq m⁻²) and geometric standard deviation 1.16 (<u>Seed et al.</u> [1971] indicated an average value of 2.4 mCi km⁻² [89 Bq m⁻²]).

Krey and Krajewski (1972) used isotopic ratios to evaluate RFP and fallout contributions to total plutonium in soil. In October 1971, they obtained additional soil samples from locations 24 and 28 of the previous HASL sampling documented in Krey and Hardy (1970) (see Figure H-4). The new samples were taken to a depth of 10 cm (4 in.). The sample analyses were for isotopic ²³⁹Pu and ²⁴⁰Pu, in addition to total ^{239,240}Pu. Ratios of ²⁴⁰Pu to ²³⁹Pu were then calculated for the samples at locations 24 and 28, as well as for two "reference" locations known to contain primarily fallout plutonium and primarily RFP plutonium. Because the ratios for RFP plutonium and worldwide fallout plutonium were significantly different, it was possible to calculate the amounts of plutonium that originated from fallout and from the RFP for locations 24 and 28. The total measured ^{239,240}Pu concentrations at locations 24 and 28 were 2.39 (±2.5%) and 1.67 (±2.5%) mCi km⁻² (88 and 62 Bq m⁻²), respectively. For these two locations, the concentrations of ^{239,240}Pu that originated from fallout were then calculated to be 1.49 and 1.52 mCi km⁻² (55 and

56 Bq m⁻²). Thus, Krey and Krajewski estimated the background concentration of 239,240 Pu because of fallout to be 1.5 mCi km⁻² (56 Bq m⁻²). The remaining 239,240 Pu in the samples appeared to be due to releases from the RFP.

Krey (1976) applied the isotopic ratio methods of Krey and Krajewski (1972) to an expanded sampling program. In September and October 1972, soil samples were collected from previous locations 22, 23, 27, and 29–32, and from 10 new locations, 34–43 (see Figure H-4). As seen in the figure, these locations ranged from a few kilometers from the RFP to about 64 km (40 mi) from the plant. For this study, sample depth was 10 cm (4 in.), as that depth was thought to contain about 90% of the deposited plutonium. For the analysis, Krey also included the results from locations 24 and 28 from the previous study of Krey and Krajewski (1972). Total measured deposition of ^{239,240}Pu was 1.13–2.87 mCi km⁻² (41.8–106 Bq m⁻²). From the ratios of ²⁴⁰Pu to ²³⁹Pu, the ^{239,240}Pu deposition from the RFP was calculated. We performed the subtraction to obtain the estimated ^{239,240}Pu deposition from global fallout. The global fallout deposition was 1.12–2.51 mCi km⁻² (41.4–92.9 Bq m⁻²). The mean ^{239,240}Pu deposition because of global fallout was calculated by Krey to be 1.7 ±0.5 mCi km⁻² (63 ±20 Bq m⁻²). Table H-10 summarizes the estimated background concentrations of plutonium in soils based on the HASL studies.

Table H-10. Summary of Determinations of Background (Global Fallout)Total Deposition of ^{239,240}Pu in Soils within 64 Kilometers (40 Miles) of the Rocky FlatsPlant, by the Health and Safety Laboratory

		Deposition	of ^{239,240} Pu ^a		
Date	Sites	(mCi km ⁻²)	$(Bq m^{-2})$	 Determination method	Reference
1965	1	1.5	56	"Background" location	Krey and Hardy (1970)
1970	33	$2.3 \times \div 1.16^{b}$	$85 \times \div 1.16^{b}$	Log-probability analysis of distribution of results	<u>Seed et al.</u> (1971) ^c
1971	2	1.50	56	²⁴⁰ Pu: ²³⁹ Pu ratios	<u>Krey and Krajewski</u> (1972)
1972	19	1.7 ± 0.5^{d}	63 ± 20^d	²⁴⁰ Pu: ²³⁹ Pu ratios	<u>Krey</u> (1976)

^a Results were reported in source documents in units millicuries per square kilometer.

^b The $\times/$ ÷ value here is one geometric standard deviation of the samples.

^c Authors were with Dow Chemical. Estimated background was based on their analysis of HASL data.

^d The \pm value here is one standard deviation of the average.

Rocky Flats Plant Routine Sampling

Routine soil sampling for plutonium analyses has been conducted at the RFP from the 1970s. The sampling program has changed over the years, so we examine results from a few select years to determine the usefulness of the data to represent background plutonium concentrations in soil.

In 1972, 20 locations were sampled on each of three concentric rings around the RFP, at 1.6, 3.2, and 8-km (1, 2, and 5-mi) radii (Boss et al. 1973). Surface samples were collected to a depth of 5 cm (2 in.). For the 8-km (5-mi) radius ring, concentrations of 239,240 Pu were 5.2–36 Bq kg⁻¹. The uncertainties in these values were extremely large, sometimes greater than 100%.

In 1980, the locations farthest from the center of the site were three locations at the eastern boundary of the site, near Indiana Street (<u>Hornbacher et al.</u> 1981). At each location, nine

composite samples were obtained. From the 27 samples at these locations, the concentrations of 239,240 Pu were 28–150 Bq kg⁻¹.

In 1991, samples were again taken in concentric rings, although now only at 1.6 and 3.2-km (1 and 2-mi) radii (<u>Altman et al.</u> 1992). Surface samples were collected to a depth of 5 cm (2 in.). For the 3.2-km (2-m) radius ring, concentrations of ^{239,240}Pu were 0.37–130 Bq kg⁻¹. The 1991 annual report (<u>Altman et al.</u> 1992) also summarized results from 1984–1991. For the other years, some had higher maximum concentrations, and one had lower minimum concentrations.

From the results of 1984–1991, some concentrations of ^{239,240}Pu in soil were less than 4 Bq kg⁻¹ and, thus, within the range of background seen from other studies. However, none of the sampling locations were specifically chosen to represent background plutonium concentrations unaffected by releases from the RFP. In addition, analyses of the data were insufficient to determine that the measured concentrations were not influenced by plutonium from the RFP. Thus, these data from routine sampling by the RFP may not be as useful as some of the other data in determining the background concentrations. However, the lowest concentrations measured by these studies should provide an indication of background levels.

Colorado State University Study

A CSU study sampled soil extensively from around the RFP during 1992–1994 (Webb 1996). This study included 10 background locations along the front range of the Rocky Mountains, shown in Figure H-3. For the background locations, three different sampling depths were used: 0.3, 3, and 21 cm (0.12, 1.2 and 8 in.). Results of this sampling are given in Table H-11. For the 0 to 21-cm (0 to 8 in.) samples, the result of 3.27 Bq kg⁻¹ for location Z10 appeared abnormally high because results for the other locations are 0.22 to 0.62 Bq kg⁻¹. Because of the significant difference in the value for Z10 and the other values, we did not calculate a mean concentration for the 0 to 21-cm (0 to 8-in.) depth. However, there is no information to indicate that the value should be disregarded, and it is probably within the range of statistical variation.

An estimate of the background inventory (total quantity) of plutonium was also described in <u>Webb</u> (1996). Many locations (in addition to the background locations) around the RFP were sampled. At some of these locations, depth profile data were obtained by taking samples in 3-cm (1.2-in.) increments to a depth of 21 cm (8 in.). With concentrations at varying locations and depths, CSU developed models to describe the concentrations as functions of distance and direction from the 903 Area and depth in the soil. These models were then used to develop the following inventory model, which describes the total deposition of plutonium (<u>Webb</u> 1996):

$$\mathbf{I}^{\theta,D} = \left(55 \ kg \ m^{-2}\right) \left[{}^{239} Pu \right]_{0-3 \ cm}^{\theta,D} \tag{H-3}$$

where

 $I^{0,D}$ = inventory, or total deposition, of ^{239,240}Pu in soil at distance *D* and direction 2 from the 903 Area (Bq m⁻²)

 $\begin{bmatrix} 2^{39} & \text{Pu} \end{bmatrix}_{0-3 \text{ cm}}^{\theta, D} = \text{concentration of plutonium in the 0 to 3-cm layer of soil at distance } D \text{ and } direction 2 \text{ from the 903 Area (Bq kg^{-1}).}$

	Sample depth				
Location	0–0.3 cm	0–3 cm	0–21 cm		
Z01	0.86	1.20	0.22		
Z02	1.52	2.10	0.45		
Z03	1.62	1.46	0.33		
Z04	1.29	2.10	0.49		
Z04 ^b	с	с	0.46		
Z05	2.33	2.10	0.51		
Z06	0.96	1.14	0.43		
Z06 ^b	с	с	0.49		
Z07	с	3.29	с		
Z08	1.51	3.22	0.48		
$Z08^{b}$	с	с	0.62		
Z09	1.43	2.07	с		
Z10	2.47	2.70	3.27 ^d		
Mean	1.55	2.14	d		
Standard deviation	0.54	0.76	d		

Table H-11. Background (Fallout) Concentrations of
^{239,240} Pu in Colorado Soils Measured by Colorado State
University in 1992–1994 $(Bg kg^{-1})^{a}$

^a Source: Webb (1996).

^b This second result is for a split sample.

^c No sample.

The value for the 0–21 cm depth at location Z10 seems abnormally high, relative to the other locations; therefore, we did not calculate mean and standard deviation.

<u>Webb</u> (1996) used this equation to calculate the total quantity of background ²³⁹Pu in the study area (this was total activity, in giga-becquerel [GBq]). In this present report, we perform essentially the same calculation, but we only calculate the intermediate result of average background ^{239,240}Pu deposition (in becquerels per square meter). This is done by applying the equation above to the average background concentration in 0 to 3-cm (0 to .2-in.) soil. The average background concentration in 0 to 3-cm (0 to 1.2-in.) soil is 2.14 Bq kg⁻¹, as used by CSU, and as shown in Table H-11. Thus, the average background deposition is estimated to be (55 kg m⁻²) × (2.14 Bq kg⁻¹) = 118 Bq m⁻². Note that this estimate is based on measured background concentrations for 0 to 3 cm (0.1.2 in.), and on models describing the depth distribution of the plutonium in soil.

EG&G Study

A study by the RFP focused on the characterization of background soils around the RFP (<u>EG&G 1995</u>). This Background Soils Characterization Program (BSCP) included soil sampling at 50 sites remote from the RFP, all in undisturbed areas along the front range of Colorado. Samples were analyzed for concentrations of fallout radionuclides, including ^{239,240}Pu. In addition, 12 samples were subjected to isotopic analyses so that ratios of ²⁴⁰Pu to ²³⁹Pu could be determined. The sampling locations, in relation to the RFP, are shown in Figure H-5.



Figure H-5. Background sampling locations and concentrations of ^{239,240}Pu in surface soil along the front range, measured in 1994 (EG&G 1995). The left side of this figure shows the sampling locations, to scale. The right side shows the measured plutonium concentrations. For a given location, the measured concentration is shown directly to the right of the location. Data are from Table H-12.

included in the mean shown in Table H-12.

At each sampling site, two 1 m × 1 m (3.3×3.3 ft) square areas were located. From each of the two areas, 5 subsamples were taken, from each of the corners and from the center, and the 10 subsamples were composited to form the sample for analysis. Each of the subsamples was taken to a depth of 5 cm (2 in.), using a 10 cm × 10 cm (4×4 in.) square template. Table H-12 shows the results for each sampling location. For some locations, duplicate soil samples were obtained in the field or replicate analyses were performed in the laboratory. In such cases, the values shown in Table H-12 are means of the duplicate or replicate measurements. The result for location GM3 deserves further explanation. The original result for location GM3 appeared to be an outlier. The measured concentration, 0.35 pCi g⁻¹, was 4.6 times higher than the maximum of all other analyses and about 10 times higher than the mean of the other analyses. To investigate the result, two replicate analyses were performed on a duplicate field sample. The results of the four additional replicates were 0.025–0.032 pCi g⁻¹, about 10 times lower than the original result. It was concluded that the original result was likely because of a laboratory error rather than to elevated plutonium at the sampled location (EG&G 1995). Thus, the original, high result is not

Site	Concentration	Site	Concentration	Site	Concentration	Site	Concentration
AF1	1.26	DR1	0.92	JP3	2.04	RM3	1.15 ^b
AF2	1.00	DR2	1.07	LH1	1.92	RR1	1.22
AF3	0.63	DR3	0.89	MR1	0.92	RR2	1.07
BE1	1.37	ES1	2.00	MR2	2.26^{b}	TH1	1.63
BE2	1.85	ES2	0.96	MR3	1.09 ^b	TH2	0.89
BE3	1.70	ES3	1.41	MW1	0.85	TH3	1.26
CM1	1.22	FW1	0.81	MW2	0.96	TM1	2.65 ^c
CM2	2.04	FW2	1.70	PP1	1.54 ^b	TM2	1.85
CM3	0.81	GM1	2.48	PR1	1.15	TM3	1.37
CR1	0.63°	GM2	1.30	PR2	1.33	TM4	2.66
DP1	1.15	GM3	1.08^{d}	PR3	1.22	TM5	1.52
DP2	2.33	JP1	1.00	RM1	2.40		
DP3	0.78	JP2	1.22 ^c	RM2	1.26 ^b		

Table H-12. Background Concentrations of ^{239,240}Pu in Surface Soil Along the Front Range, Measured by EG&G in 1994 (Bq kg⁻¹)^a

^a Measurements have been converted from units of picocuries per gram , in the original reference (EG&G <u>1995</u>), to becquerels per kilogram.

^b Mean of results of two replicate analyses.

^c Mean of results of duplicate soil samples.

^d Mean of results of two replicate analyses of each of two duplicate soil samples. Original result, which was determined to be erroneous, is not included (see text for details).

The results of the background concentrations from the BSCP are also shown in Figure H-5, which depicts the plutonium concentrations in relation to the sampling locations. From the 50 sampling locations, the concentrations of 239,240 Pu ranged from 0.629 to 2.664 Bq kg⁻¹. The mean concentration was 1.40 Bq kg⁻¹, and the standard deviation was 0.54 Bq kg⁻¹. One important conclusion from Figure H-5 is that there does not appear to be any significant trend in plutonium concentration with distance from the RFP, which indicates that the sampling locations probably

represent background concentrations not affected by releases from the RFP. From the 12 samples analyzed isotopically, the mean 240 Pu/ 239 Pu ratio was 0.1552, and the standard deviation was 0.0093 (EG&G 1995).

Summary of Background Plutonium in Soil near the Rocky Flats Plant

In summary, the measurements performed by NCAR, CDH, CSU, and EG&G (Tables H-7, H-8, H-9, H-11, and H-12) indicate that the background mass concentration of 239,240 Pu in surface soils (0.16 to 5-cm [0.06 to 2-in.] depths) of eastern Colorado is in the range of about 0.3–4.5 Bq kg⁻¹ (0.008–0.1 pCi g⁻¹), although only one value was greater than 3.3 Bq kg⁻¹. The wide variability in results may be due to the very shallow surface layers of soil that were sampled and the spatial patterns of fallout deposition across the large area covered by sampling. To summarize the deposition measurements and calculations performed by HASL (and the analysis of HASL results by Seed et al. [1971]) and by CSU we used the values from Tables H-10 and H-11. For the HASL values, we also included one standard deviation or one geometric standard deviation (where available) to represent likely ranges (Table H-10). These results indicate that the total deposition of 239,240 Pu from global fallout, in the general area around the RFP and along the front range, was probably in the range of 40–120 Bq m⁻² (1.1–3.2 mCi km⁻²).

BACKGROUND PLUTONIUM IN SOIL AT GREATER DISTANCES FROM THE ROCKY FLATS PLANT

This section describes measurements of background soil concentrations of plutonium for locations farther from the RFP. While concentrations at great distances from the RFP may not be indicative of the background around the plant, they do provide some perspective as to how local background concentrations compare with regional and global background.

Purtymon et al. (1990) reports data on soil concentrations of plutonium in northern New Mexico and southern Colorado, which are in the same general region as the RFP, and within 5° latitude. In this study, six locations were sampled in 1981 and 1983, and nine separate locations were sampled in 1986. The locations were all east of (or on) the continental divide. The northernmost location was Monarch Pass, Colorado, about 160 km (100 mi) from the RFP, and the southern-most location was Santa Ana Pueblo, New Mexico, about 480 km (300 mi) from the RFP. Some of the locations are, however, within about 32 km (20 mi) of the Los Alamos National Laboratory, which is a potential source of plutonium in the environment. The soil samples were composites made up of five subsamples, taken to a depth of 5 cm (2 in.). Alpha spectroscopy measurements were performed to obtain ²³⁸Pu and ^{239,240}Pu, which were summed to obtain total plutonium. On the average, ²³⁸Pu contributed less than 5% to the total plutonium activity measured. We only consider the ^{239,240}Pu measurements here for comparability with other measurements. Concentrations of ^{239,240}Pu ranged from 1.2 to 81 fCi g⁻¹ (0.044 to 3.0 Bq kg⁻¹), with an average of 14 fCi g^{-1} (0.53 Bq kg⁻¹) and a standard deviation of 18 fCi g^{-1} (0.66 Bq kg⁻¹). The two highest values occurred for locations in high mountain passes on the continental divide. As discussed earlier, higher values are expected for high mountain areas, and it may be reasonable to consider these locations grouped separately from the remaining locations. If these highest values are disregarded, the remainder cover the range of 1.2-19 fCi g⁻¹ (0.044-0.71

Bq kg⁻¹), with an average of 9.4 fCi g⁻¹ (0.35 Bq kg⁻¹) and a standard deviation of 5.4 fCi g⁻¹ (0.20 Bq kg⁻¹).

As mentioned above, <u>Holleman et al.</u> (1987) provides an extensive compilation of datasets on worldwide fallout of plutonium from weapons tests. From this compilation, we extracted measured concentrations of ^{239,240}Pu in soil in the U.S. Values were given in units of total deposition (becquerels per square meter) and mass concentration (becquerels per kilogram). Information about individual measurements is given in Table H-16, at the end of this appendix. Holleman et al. does not provide information about sample depths, but this is not necessary for our work. The time the samples were collected is not that important because the earliest date (1962) is after the majority of plutonium was deposited (Figure H-2). The results are summarized by state in Table H-13; for some states only one measurement was available.

 Table H-13. Summary of ^{239,240}Pu Deposition and Mass Concentrations in Soil in the United States (from the Compilation by <u>Holleman et al. 1987</u>)

				Deposition (Bq m ⁻²) ^a		Concer	ntration (Bq	$(kg^{-1})^a$	
State	Sites	Samples	Dates	Min	Max	Avg	Min	Max	Avg
Alaska	5	9	1964–1976	1.18	34	13			
California	3	5	1970-1972	27	37	32			
Colorado	1	7	1965-1970	2.11	67	47			
Florida	1	1	1970			37			
Hawaii	1	1	1970			148			
Illinois	17	62	1970-1981	10.36	256.78	51			
Kansas	1	1	1970			89			
Maine	1	1	1970			63			
Massachusetts	1	1	1972			85			
Michigan	1	1	1976			99.9			
Montana	1	1	1965			70			
New Mexico	6	36	1974–1977				0.0	0.78	0.28
New York	7	16	1964-1973	67	99.9	84			
North Carolina	1	1	1970			89			
Ohio	1	25	1974				0.114	1.528	0.28
Oklahoma	1	1	1970			81			
South Dakota	2	2	1965-1970	85	93	89			
Texas	2	2	1970	32.6	36.6	35			
Utah	1	1	1970			96			
Washington	2	7	1970-1971	1.5	52	20			
Wisconsin	1	1	1972			58			

^a Min = minimum, max = maximum, and avg = arithmetic average. Minimum and maximum values are taken from the source document (<u>Holleman et al. 1987</u>), and the averages are calculated by us, in this present work. The values presented retain the number of significant figures used by Holleman et al., though we acknowledge that in some cases they are excessive.

The samples in Ohio were taken near the Mound facility, which processed plutonium; however, this facility handled primarily ²³⁸Pu. The very low values of deposition reported for Alaska are probably because of the more northerly latitude of Alaska. The single result for Hawaii is relatively high compared to other states closest in latitude (Florida and Texas). The elevated value may be due to Hawaii's proximity to some of the weapons tests in the Pacific and, thus, may reflect some regional (in addition to global) fallout. The minimum deposition value for Colorado (2.11 Bq m⁻²) does not appear to be a credible value; deposition this small seems

Deposition ^a			Concentration ^a							
Latitude band	Sites	Samples	Min	Max	Avg	Sites	Samples	Min	Max	Avg
<32.5 °N ^b	3	3	33	37	35					
32.5–37.5 °N	5	7	27	89	47	6	36	0	0.78	0.28
37.5-42.5 °N ^c	30	91	10	260	60	1	25	0.11	1.5	0.28
42.5–47.5 °N	5	10	1.5	93	37					
>47.5 °N ^d	5	9	1.2	34	13					

Table H-14. Summary of ^{239,240}Pu Deposition (Bq m⁻²) and Mass Concentrations (Bq kg⁻¹) in Soil in the United States, by Latitude (from the Compilation in <u>Holleman et al. 1987</u>)

^a Min = minimum, max = maximum, and avg = arithmetic average. Minimum and maximum values are taken from the source document (<u>Holleman et al. 1987</u>), and the averages are calculated by us, in this present work.

^b The single result for Hawaii is not included in this summary, because the value is probably not representative of *global* fallout.

^c The extremely low value from Colorado is not included in this summary.

^d All values in this latitude band are from Alaska, at latitudes greater than 60 °N.

extremely unlikely. However, the average value for Colorado is in reasonable agreement with other states at similar latitudes (e.g., Illinois).

Because global fallout deposition is correlated with latitude in the northern hemisphere, we also summarized the ^{239,240}Pu measurements by latitude bands. We centered the bands around latitude 40 °N because that is the approximate location of the RFP. Table H-14 shows this summary.

CONCLUSIONS ON BACKGROUND PLUTONIUM IN SOILS

From the studies presented here, the measured levels of plutonium in soil around the RFP and around the U.S. are compared in Table H-16. (We acknowledge that this is not necessarily a complete compilation of such data.) Figure H-6 compares the total plutonium deposition for background locations around the RFP to locations in the U.S. at similar latitudes. The range of measured deposition of 239,240 Pu around the RFP (40–120 Bq m⁻², or 1.1–3.2 mCi km⁻²) is within that seen for other states in the 37.5–42.5 °N latitude range (10–260 Bq m⁻², or 0.27–7 mCi km⁻²), although they tend slightly toward the higher end of measured concentrations. The measured mass concentrations of 239,240 Pu around the RFP (<0.3–4.5 Bq kg⁻¹, or 0.008–0.1 pCi g⁻¹) exceed (slightly) the ranges of values seen in New Mexico and Ohio measurements (0–3.0 Bq kg⁻¹, or 0–0.08 pCi g⁻¹). Many of the lowest values for the U.S. are for locations, such as Alaska, not in the middle latitudes. Thus, it appears that while measured background concentrations of plutonium in soil around the RFP tend to be higher than background concentrations for many locations, they are still within the ranges observed in other states at similar latitudes.

Some important characteristics related to plutonium in soils should be considered in evaluations of soil sample results around the RFP. The measurement technique, and more specifically the plutonium isotopes actually measured, should be determined. Essentially all measurements of "²³⁹Pu" are actually measurements of ^{239,240}Pu because alpha spectroscopy is commonly used for the analyses. If isotopic results, such as the ratio ²⁴⁰Pu/²³⁹Pu, are available, it may be feasible to determine more accurately whether the source of the plutonium is truly background or if it has been influenced by RFP sources. When comparing samples near the RFP

to background samples, the time of sample collection can be important because there are temporal trends in the global fallout of plutonium from nuclear weapons testing. Finally, depth distributions of plutonium should be considered relative to the goals of a particular sampling program or analysis. Soil samples taken from the surface soils (e.g., to 1 cm [0.4 in.] or so) are generally not representative of the total deposition of plutonium that exists in the soil column. Quantitative comparisons between results of sampling programs with widely disparate sample depths should be performed when information is available to develop relationships between soil layers of different depths.

	Deposition (Bq m^{-2})		Concentrati	ion (Bq kg ⁻¹)
Locations	Minimum	Maximum	Minimum	Maximum
Around RFP	40	120	< 0.3	4.5
37.5–42.5 °N Latitude ^a	10	260	0.11	1.5
U.S.	1.2	260	0.0^{b}	3.0^{b}

 Table H-15. Comparison of Measured Deposition and Mass Concentrations of ^{239,240}Pu

 Around the Rocky Flats Plant with those Around the United States

^a The extremely low value from Colorado is not included in this summary.

^b Mass concentrations were from two states only: New Mexico and Ohio.



Figure H-6. Background ^{239,240}Pu total deposition (inventory) in soils: comparison of levels around the RFP with levels in the U.S. at similar latitude. Data are described in this appendix. Notes: (a) single location, in 1965, (b) examined distribution of measurements from 1970, (c) used ²⁴⁰Pu/²³⁹Pu ratios for samples from 1971, (d) used ²⁴⁰Pu/²³⁹Pu ratios for samples from 1972, (e) samples from 1992–1994, 0–3-cm (0–1.2-in.) depth, with depth distribution model, (f) from compilation of numerous measurements.

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ANNEX

				^{239,240} Pu co	ncentration
State	Location	Latitude (°N)	Date	$Bq m^{-2}$	$Bq kg^{-1}$
Alaska	Anaktuvuk Pass	68.10	Jul 1975	5.62	
Alaska	Anaktuvuk Pass	68.10	Jul 1976	1.55	
Alaska	Anaktuvuk Pass	68.10	Sep 1976	1.18	
Alaska	Barrow	71.17	Aug 1964	12.20	
Alaska	Barrow	71.17	1970	14.8	
Alaska	Bettles	66.55	Jul 1976	4.26	
Alaska	Fairbanks	64.51	Jul 1976	8.21	
Alaska	Fairbanks	64.51	1970	31.4	
Alaska	Palmer	61.36	1970	34	
California	Burbank	34.2 ^b	1970	27	
California	Oakland	37.47	Oct 1972	30.00	
California	Oakland	37.47	Oct 1972	30.00	
California	San Francisco	37.48	Oct 1972	34.00	
California	San Francisco	37.48	Oct 1972	37.00	
Colorado	Denver	39.43	Sep 1965	56	
Colorado	Denver	39.43	Feb 1970	32.9	
Colorado	Denver	39.43	Feb 1970	40.7	
Colorado	Denver	39.43	Sep 1970	65	
Colorado	Denver	39.43	Sep 1970	2.11	
Colorado	Denver	39.43	Oct 1970	67	
Colorado	Denver	39.43	1970	67	
Florida	Ft. Pierce	27.27	1970	37	
Hawaii	Papaikou	19.47	1970	148	
Illinois	Argonne	41.43	1970	78	
Illinois	Brookfield	41.49	Sep 1972	57.35	
Illinois	Brookfield	41.49	Oct 1974	65.86	
Illinois	Brookfield	41.49	Jun 1976	70.3	
Illinois	Brookfield	41.49	Jun 1979	36.63	
Illinois	Brookfield	41.49	Oct 1980	49.21	
Illinois	Channahon	41.26	Jun 1978	49.6	
Illinois	Channahon	41.26	Jun 1979	31.08	
Illinois	Channahon	41.26	Jun 1980	19.61	
Illinois	Channahon	41.26	Jun 1981	25.2	
Illinois	Downers Grove	41.48	Jun 1979	18.5	
Illinois	Downers Grove	41.48	Oct 1981	29.2	
Illinois	Dresden Lock and Dam	41.20	Oct 1976	74	
Illinois	Dresden Lock and Dam	41.20	Oct 1978	45.1	
Illinois	Dresden Lock and Dam	41.20	Oct 1979	15.91	
Illinois	Dresden Lock and Dam	41.20	Oct 1980	10.36	
Illinois	Dresden Lock and Dam	41.20	Oct 1981	41.8	
Illinois	Hinsdale	41.48	Oct 1974	127.65	
Illinois	Hinsdale	41.48	Jun 1976	81.4	
Illinois	Lemont	41.40	Nov 1974	56.61	
Illinois	Lemont	41.40	Oct 1978	21.5	
Illinois	Lemont	41.40	Jun 1980	19.61	
Illinois	Lemont	41.40	Oct 1981	23.7	

Table H-16. Individual Measurements of ^{239,240}Pu in Soil in the United States(from the Compilation of Holleman et al. 1987)^a

				^{239,240} Pu co	ncentration
State	Location	Latitude (°N)	Date	Bq m^{-2}	$Bq kg^{-1}$
Illinois	McGinnis Slough	41.39	Sep 1972	72.52	
Illinois	McGinnis Slough	41.39	May 1974	80.3	
Illinois	McGinnis Slough	41.39	Oct 1978	42.5	
Illinois	McGinnis Slough	41.39	Jun 1980	22.57	
Illinois	Mckinley Woods State Park	41.45	Jun 1972	40.7	
Illinois	Mckinley Woods State Park	41.45	Oct 1974	77.7	
Illinois	Mckinley Woods State Park	41.45	Jun 1976	114.7	
Illinois	Mckinley Woods State Park	41.45	Oct 1978	54.8	
Illinois	Mckinley Woods State Park	41.45	Oct 1979	35.52	
Illinois	Mckinley Woods State Park	41.45	Oct 1980	20	
Illinois	Mckinley Woods State Park	41.45	Oct 1981	69.2	
Illinois	Morris	41.22	May 1974	75.85	
Illinois	Morris	41.22	May 1974	256.78	
Illinois	Morris	41.22	Jun 1978	52.2	
Illinois	Morris	41.22	Jun 1979	32.56	
Illinois	Morris	41.22	Jun 1980	27	
Illinois	Morris	41.22	Jun 1981	17	
Illinois	Naperville	41.47	Jun 1972	55.5	
Illinois	Naperville	41.47	May 1974	94	
Illinois	Naperville	41.47	Jun 1978	57.7	
Illinois	Naperville	41.47	Jun 1981	24	
Illinois	Romeoville	41.39	Oct 1978	57.3	
Illinois	Romeoville	41.39	Oct 1981	44.4	
Illinois	Saganashkee Slough	41.41	Jun 1972	77.33	
Illinois	Saganashkee Slough	41.41	May 1974	72.52	
Illinois	Saganashkee Slough	41.41	Oct 1978	27	
Illinois	Saganashkee Slough	41.41	Jun 1980	21.83	
Illinois	Starved Rock State Park	41.19	May 1974	76.22	
Illinois	Starved Rock State Park	41.19	Jun 1978	43.3	
Illinois	Starved Rock State Park	41.19	Jun 1979	31.08	
Illinois	Starved Rock State Park	41.19	Jun 1980	17.39	
Illinois	Starved Rock State Park	41.19	Jun 1981	43.7	
Illinois	Western Springs	41.47	Jun 1979	35.9	
Illinois	Western Springs	41.47	Oct 1980	24.8	
Illinois	Willow Springs	41.50	Oct 1976	107.3	
Illinois	Willow Springs	41.50	Jun 1978	27	
Illinois	Willow Springs	41.50	Oct 1979	30.71	
Illinois	Woodridge	41.46	Oct 1979	32.56	
Illinois	Woodridge	41.46	Iun 1981	30.3	
Kansas	Manhattan	39.11	1970	89	
Maine	Orono	44.53	1970	63	
Massachusetts	North Eastham, Cape Cod	41.52	Oct 1972	85	
Michigan	St Joseph	42.06	Oct 1976	99.9	
Montana	Bozeman	45 41	Sep 1965	70	
New Mexico	Bernalillo	35.30	Jul 1974	10	0.22
New Mexico	Bernalillo	35 30	May 1975		0.44
New Mexico	Bernalillo	35 30	Oct 1975		0.04
New Mexico	Bernalillo	35 30	Apr 1976		0.15
New Mexico	Bernalillo	25 20	Oct 1076		0.07
New Mexico	Bernalillo	35.30	Mar 1077		0.07
New Mexico	Bernalillo	25 20	Oct 1077		0.07
New Mexico	Chamita	36.00	Jul 1974		0.22
THE MILLION	Challinta	50.00	Jul 17/T		0.22

Table H-16. Individual Measurements of ^{239,240}Pu in Soil in the United States, from the Compilation of Holleman et al. (1987) (continued)

				^{239,240} Pu co	oncentration
State	Location	Latitude (°N)	Date	Bq m ⁻²	Bq kg ⁻¹
New Mexico	Chamita	36.00	Oct 1975		0.63
New Mexico	Chamita	36.00	Mar 1976		0.3
New Mexico	Chamita	36.00	Oct 1976		0.52
New Mexico	Chamita	36.00	Mar 1977		0.63
New Mexico	Chamita	36.00	Oct 1977		0.37
New Mexico	Cochiti	35.45	May 1975		0.07
New Mexico	Cochiti	35.45	Oct 1975		0
New Mexico	Cochiti	35.45	Apr 1976		0.15
New Mexico	Cochiti	35.45	Oct 1976		0.11
New Mexico	Cochiti	35.45	Mar 1977		0.04
New Mexico	Cochiti	35.45	Oct 1977		0.11
New Mexico	Embudo	36.00	Jul 1974		0.19
New Mexico	Embudo	36.00	May 1975		0.3
New Mexico	Embudo	36.00	Oct 1975		0.33
New Mexico	Embudo	36.00	Mar 1976		0.44
New Mexico	Embudo	36.00	Oct 1976		0.7
New Mexico	Embudo	36.00	Mar 1977		0.4
New Mexico	Embudo	36.00	Oct 1977		0.56
New Mexico	Jemez	35.45	Jul 1974		0.04
New Mexico	Jemez	35.45	May 1975		0.04
New Mexico	Jemez	35.45	Sep 1975		0.44
New Mexico	Jemez	35.45	Apr 1976		0.07
New Mexico	Jemez	35.45	Oct 1976		0.26
New Mexico	Jemez	35.45	Mar 1977		0.7
New Mexico	Jemez	35.45	Oct 1977		0.04
New Mexico	Otowi	35.50	Jul 1974		0.44
New Mexico	Otowi	35.50	May 1975		0.22
New Mexico	Otowi	35.50	Oct 1977		0.78
New York	Bronx	40.49	Jul 1970	92.5	
New York	Bronx	40.49	Aug 1970	81.4	
New York	Brookhaven National Laboratory	40.54	Sep 1970	96	
New York	Brookhaven National Laboratory	40.54	Sep 1970	78	
New York	Brookhaven National Laboratory	40.54	Nov 1972	99.9	
New York	Brookhaven National Laboratory	40.54	Nov 1972	91.8	
New York	Brookhaven National Laboratory	40.54	Nov 1972	90.6	
New York	Brookhaven National Laboratory	40.54	Nov 1972	81	
New York	Brookhaven National Laboratory	40.54	1972	88.8	
New York	Brooklyn	40.42	Nov 1972	78	
New York	Fordham University	40.51	Dec 1969	74	
New York	Fordham University	40.51	Jan 1970	81.4	
New York	Fordham University	40.51	Jan 1970	96	
New York	Kitchawan, Westchester County	41.15	Jun 1973	70.3	
New York	New York	40.43	Dec 1964	67	
New York	New York	40.43	1970	96	
New York	Teatown, Westchester County	41.15	Jun 1973	70.3	
North Carolina	Raleigh	35.47	1970	89	
Ohio	Miamisburg	39.38	Oct 1974		0.177
Ohio	Miamisburg	39.38	Oct 1974		0.222
Ohio	Miamisburg	39.38	Oct 1974		0.166
Ohio	Miamisburg	39.38	Oct 1974		0.269
Ohio	Miamisburg	39.38	Oct 1974		0.206
Ohio	Miamisburg	39.38	Oct 1974		0.171

Table H-16. Individual Measurements of ^{239,240}Pu in Soil in the United States, from the Compilation of Holleman et al. (1987) (continued)

				^{239,240} Pu co	oncentration
State	Location	Latitude (°N)	Date	Bq m ⁻²	$Bq kg^{-1}$
Ohio	Miamisburg	39.38	Oct 1974		0.256
Ohio	Miamisburg	39.38	Oct 1974		0.171
Ohio	Miamisburg	39.38	Oct 1974		0.2
Ohio	Miamisburg	39.38	Oct 1974		0.129
Ohio	Miamisburg	39.38	Oct 1974		0.17
Ohio	Miamisburg	39.38	Oct 1974		0.135
Ohio	Miamisburg	39.38	Oct 1974		0.207
Ohio	Miamisburg	39.38	Oct 1974		0.114
Ohio	Miamisburg	39.38	Oct 1974		0.174
Ohio	Miamisburg	39.38	Oct 1974		0.191
Ohio	Miamisburg	39.38	Oct 1974		0.179
Ohio	Miamisburg	39.38	Oct 1974		0.18
Ohio	Miamisburg	39.38	Oct 1974		0.19
Ohio	Miamisburg	39.38	Oct 1974		0.14
Ohio	Miamisburg	39.38	Oct 1974		0.213
Ohio	Miamisburg	39.38	Oct 1974		0.208
Ohio	Miamisburg	39.38	Oct 1974		0.16
Ohio	Miamisburg	39.38	Oct 1974		1.214
Ohio	Miamisburg	39.38	Oct 1974		1.528
Oklahoma	Tulsa	36.09	1970	81	
South Dakota	Rapid City	44.05	Sep 1965	93	
South Dakota	Vermillion	42.47	1970	85	
Texas	Kingsville	27.31	1970	36.6	
Texas	Weslaco	26.09	1970	32.6	
Utah	Salt Lake City	40.46	1970	96	
Washington	Hanford Reservation	46.50 ^b	Feb 1971	19.20	
Washington	Hanford Reservation	46.50^{b}	Feb 1971	28.10	
Washington	Hanford Reservation	46.50^{b}	Feb 1971	24.00	
Washington	Hanford Reservation	46.50 ^b	Feb 1971	8.1	
Washington	Hanford Reservation	46.50 ^b	Feb 1971	1.5	
Washington	Hanford Reservation	46.50 ^b	Feb 1971	7.8	
Washington	Puyallup	47.11	1970	52	
Wisconsin	Lake Delavan	42.38	Oct 1972	58.46	

Table H-16. Individual Measurements of ^{239,240}Pu in Soil in the United States, from the Compilation of Holleman et al. (1987) (continued)

а

The values presented here retain the number of significant figures used by Holleman et al. (1987). The latitude given in Holleman et al. (1987) appeared to be an error. We have estimated the latitude from maps, and bshow the estimated latitude here.

Multiply value with units of:	by:	To obtain value with units of:
	Units of activity	
dpm ^a	0.0167	Bq
dpm	0.45	pCi
pCi	0.037	Bq
pCi	2.22	dpm
pCi	1000	fCi
fCi	$3.7 imes10^{-5}$	Bq
fCi	0.001	pCi
Ci	37	GBq
Bq	60	dpm
Bq	27	pCi
Bq	27,000	fCi
	Concentration units: activity per mass	
dpm g^{-1}	16.7	$Bg kg^{-1}$
$dpm g^{-1}$	0.45	$pCi g^{-1}$
$pCi g^{-1}$	37	$Bq kg^{-1}$
pCi g ⁻¹	2.22	$dpm g^{-1}$
fCi g ⁻¹	0.037	$Bq kg^{-1}$
$Bq kg^{-1}$	0.060	$dpm g^{-1}$
$Bq kg^{-1}$	0.027	$pCi g^{-1}$
$Bq kg^{-1}$	27	fCi g ⁻¹
	Concentration units: activity per area	
mCi km ^{-2}	37	Bq m^{-2}
$Bq m^{-2}$	0.027	$mCi km^{-2}$
C	oncentration units: activity per liquid volur	ne
pCi L ⁻¹	0.037	$Bq L^{-1}$
$pCi L^{-1}$	1000	$fCiL^{-1}$
$fCi L^{-1}$	0.001	pCi L^{-1}
$dpm L^{-1}$	0.0167	$\mathbf{Bq} \mathbf{L}^{-1}$
$dpm L^{-1}$	450	$fCiL^{-1}$
$\operatorname{Bq} \operatorname{L}^{-1}$	27	pCi L^{-1}
$\operatorname{Bq} \operatorname{L}^{-1}$	27,000	$fCi L^{-1}$

 Table H-17. Units Conversion Factors

^a dpm = disintegrations per minute.

Example: The following is an example of using this table of units conversion factors. If you have a value of 120 Bq kg⁻¹ and wish to convert to units of pCi g⁻¹, look in the first column to find the units you have (Bq kg⁻¹). Look in the third column to find the row that contains the units to which you want to convert (pCi g⁻¹). Use the conversion factor in the second column that corresponds with that row. In this example, that row of the table shows a factor of 0.027. Multiply the starting value (120) by 0.027 ($120 \times 0.027 = 3.24$). Thus, our starting value of 120 Bq kg⁻¹ is equal to 3.2 pCi g⁻¹, with rounding.