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Historical Review of Long-Term Soil Sampling for Environmental Surveillance at the Hanford Site and Vicinity

K. R. Price
W. H. Rickard

August 1997

Prepared for the U.S. Department of Energy
under Contract DE-AC06-76RLO 1830

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Summary

Soil samples have been collected routinely from the environs of the Hanford Site and analyzed since 1971. Correct interpretation of results depends on samples being collected from the same locations, the locations remaining relatively undisturbed, and collection and analytical procedures remaining the same or being equivalent. Historical files, documents, and annual environmental reports were reviewed to evaluate these factors. It was determined that 20 soil sampling locations, 11 onsite and 9 offsite, were established between 1971 and 1977 and represent long-term sampling locations. Sample collection and analytical procedures have remained essentially the same since 1971. The physical and ecological attributes of each long-term soil sampling location were evaluated.

During the review of historical records, a few results for 1970, 1971, and 1972 were noted as previously unreported in annual or special reports. These results are included in Appendix A. To complete the record, results previously reported in annual environmental reports are given in Appendix B. Global Positioning System (GPS) readings for 20 long-term soil sampling locations are provided in Appendix C.

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1.0 Introduction

Radioactive materials have been released into the atmosphere from some facilities at the Hanford Site since operations began in the early 1940s. Materials released into the atmosphere are eventually deposited onto the soil surface. Soil sampling has been conducted routinely since 1971 in both the onsite and offsite environments at Hanford. One of the stated purposes of soil sampling is to detect the presence and long-term buildup of long-lived radioactive materials. This objective can be attained only when samples are collected periodically from the same locations, when the soils remain relatively undisturbed, and when samples are collected and analyzed using the same or equivalent procedures. A review of these factors is the principal subject of this report. Previous reviews have addressed the first 5 years of soil sampling (Miller et al. 1977), reviewed soil sampling results through 1987 (Price 1988), and provided a statistical evaluation of soil sampling results for the period 1983 through 1993 (Poston et al. 1995).

Soil sampling outside the boundaries of Hanford operational areas has been the responsibility of the Pacific Northwest National Laboratory (PNNL)^(a) since routine sampling began in 1971. Environmental surveillance (monitoring) personnel collected the samples, and results were summarized and reported in annual environmental reports. Samples were analyzed initially by the U.S. Testing Co., Inc., a government contractor and subsequently a subcontractor to PNNL, and later by Quanterra Environmental Services, Inc., a subcontractor to PNNL. Analytical results were retained in computerized data bases as well as recorded in handwritten logbooks for backup. The locations of sampling sites and the procedures for collecting and analyzing samples have been documented since the late 1970s. The first part of this report presents the results of a review of environmental monitoring program historical files, documents, and annual environmental reports. The second part provides a review and current evaluation of the physical and ecological attributes of each long-term soil sampling location.

(a) Formerly the Pacific Northwest Laboratory (PNL), operated for the U.S. Department of Energy by Battelle Memorial Institute.

2.0 Historical Records

Environmental monitoring program historical files, documents, and annual environmental reports were reviewed for information on the following issues:

- Have soil sampling locations been relocated over the years (i.e., have sampling sites been moved a considerable distance while retaining the same literal names and/or location identifying numbers)?
- Have soil sampling procedures changed significantly over the years (i.e., have larger or smaller samples been collected or have samples been collected deeper or shallower compared to current techniques)?
- Have significant changes occurred over the years in analytical procedures used to detect plutonium in soil samples or to report data (i.e., have changes occurred in analytical procedures or data reporting techniques that could influence the interpretation of results for plutonium detected in soil samples)?
- Have all significant results for plutonium and other important long-lived radionuclides detected in soil samples been reported in annual or special reports (i.e., have all results been reported relevant to the interpretation of plutonium and other important long-lived radionuclides occurring in soil samples collected offsite)?

2.1 Records Reviewed

An effort was made to find as many historical records as possible regarding soil sampling, analyses, and results. Records were reviewed to identify historical sampling locations, the reasons for sampling, analytical results, and any other relevant information. A search for results from plutonium analyses was emphasized because of the long-lived nature of most plutonium isotopes, but results for strontium-90 and cesium-137 were noted also.

The following records were located and reviewed:

- Memos, letters, and miscellaneous papers in the archives of historical records for the environmental monitoring program
- "Specials" - notebooks in the archives of historical records containing information and results for special purpose samples collected from 1967 through 1974
- "Soil, Sediment and Vegetation," a handwritten logbook of results and information on sampling from 1971 through 1989 in the archives of historical records

- Documentation about historical computer systems and isotope identification codes located in a box of records retrieved from storage
- A copy of the original *Environmental Sampling Locations Manual* (Blummer 1979), and subsequent versions (PNNL 1983; PNNL 1991)^(a)
- Sample collection procedures found in *Environmental Monitoring Procedures* (Blummer 1981), a subsequent revision (Price et al. 1986, Hanf 1995)^(a)
- Master schedules and annual reports for 1971 and subsequent years.

2.2 Sampling Locations

The name of one sampling location in the historical record was noted as representing a different location than is in use today. A sampling site known as the "Yakima Barricade" was originally located at the site boundary on State Route 24, 3.3 miles west of the present Yakima Barricade. It was also known as the "West Intersection of Rt. No. 24 and Plant Boundary." However, this location was used before routine soil sample collection was established in 1971. The current Yakima Barricade soil sampling site is located a few hundred yards north of the Hanford Site Yakima Barricade guard house, near the existing air sampling station.

Minor adjustments to literal names have also been made over the years. For example, "ALE," "East of Arid Land Ecology Lab," and "ALE Field Lab;" "0.5 Mile NE of FFTF," "NE FFTF," and "400E;" and others. A complete list of also known as (a.k.a.) names is provided in Section 3.0, "Evaluation of Soil Sampling Locations."

More important, however, is whether or not routine soil sampling locations were changed after 1971 without changing the literal names. The use of location names was traced by examining the computer systems used to manage information for the environmental monitoring program and the various types of documentation recorded for those systems.

Since 1971, sampling locations have been mostly standardized, and analytical results have been stored in information data bases of computer systems. Several computer systems have been used since the inauguration of RMIS (Regional Monitoring IBM System) in 1956. In 1971, the EMA (Environmental Monitoring Analysis)^(b) computer system was established. The EMA system was later replaced by HEDS (Hanford Environmental Data System) and PDMS (Project and Data Management System). The HEIS

(a) Locations manuals and procedures manuals are reviewed annually and revised as necessary.

(b) A definition of the acronym EMA, "Environmental Monitoring Analyses," apparently was first recorded in 1977 in a technical procedures document prepared for the Energy Research and Development Administration (PNNL 1977).

computer system (Hanford Environmental Information System) is now in use for all environmental data collected by PNNL for the Hanford Site. All current and historical data were transferred into each new data management system when it was put into use.

2.2.1 Location Identifying EMA Numbers

Sample location identifying numbers, i.e., EMA numbers^(a), were established in 1971 for the original EMA computer system and were used to identify specific sampling locations where soil and other types of samples were collected (Blummer and Corley 1971). An apparent original EMA number logbook was found in storage box #134924^(b). The logbook identified EMA numbers for soil sampling locations in use until 1989 when data were transferred to the HEIS computer system. Although undated, the earliest handwritten entries in the logbook must have been prepared in the early 1970s (probably 1971) because soil samples were identified with EMA numbers for "soil sample 0-1 in." and "soil sample 1-2 in." Samples from each of these 1-inch-deep layers of soil were collected for the routine environmental monitoring program only in 1971 and 1972.

An original carbon copy of an "Environmental Monitoring Trip Log" was found in an historical file. The trip log (a.k.a. trip sheet) listed the EMA numbers and associated literal names for locations of the first soil samples collected in 1971 for the routine environmental monitoring program. These numbers were the same as those listed in the EMA number logbook. The trip log was dated 4-6-72, but the sample collection dates were recorded as 9-29-71, 9-30-71, and 10-1-71. An "Environmental Monitoring Analysis Report" (i.e., computerized result report) from U.S. Testing Co., Inc. dated 5-26-72 reported plutonium-238 and plutonium-239,240 data for soil samples with the same EMA numbers as indicated on the trip log dated 4-6-72. The "Soil, Sediment and Vegetation" logbook of handwritten analytical results contained data reported from 1971 to 1989. The EMA numbers for soil samples collected from 1971 through 1992 were the same as for those found in the records listed above, and they did not change over the years. Additional EMA numbers were assigned as new locations were established, but the same EMA number was not assigned to more than one location. After 1992, the HEIS computer system and its unique numbering system was put into use to maintain the identity of sampling locations.

In summary, EMA numbers were assigned to sampling locations in order to process information in the EMA computer system and subsequent systems. Specific EMA numbers for soil sampling locations were first assigned in 1971. Records show that the EMA numbers assigned to soil sampling locations established in 1971 and later years remained unchanged until a different computer system was put into use in 1992 to maintain the identity of sampling locations.

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- (a) EMA Numbers were specific for location, media, and sample type.
(b) Retrieved from Hanford Sitewide Records Services, U.S. Department of Energy, Richland, Washington.

2.2.2 Locations Manuals

A locations manual for environmental sampling was prepared in 1978 and updated in 1979 to include soil sampling locations (Blummer 1979). The 1979 locations manual contained maps, photographs, and general descriptions of all soil sampling locations in use at the time. The 1979 locations manual, and subsequent revisions published in 1983 (PNNL 1983) and in 1991 (PNNL 1991), provided formalized documentation of where samples were to be collected for the routine environmental monitoring program. A review of the three locations manuals showed the following breakdown in the number of onsite and offsite soil sampling locations:

- 1979, 11 onsite locations and 10 offsite locations

<u>Onsite</u>	<u>Offsite</u>
Wahluke Slope No. 2	Benton City
Yakima Barricade	Sunnyside
East of 200 West Area	Harris Farm
200 ENC	Byers Landing
200 ESE	Sagemoor
Hanford Townsite	Fir Road
SE FFTF	Taylor Flats No. 1
NE FFTF	Taylor Flats No. 2
Wye Barricade	Ringold
Prosser Barricade	Berg Ranch
ALE	

- 1983, 11 original plus 12 additional onsite locations (23 total), and 9 of the original offsite locations (Taylor Flats No. 1 deleted)
- 1991, the same 23 onsite locations and 9 offsite locations listed in 1983 plus 7 additional offsite locations (total 16).

Sampling was discontinued at the Taylor Flats #1 location when the access road was blocked by landslides. Thus, the original 20 soil sampling locations listed in the 1979 locations manual, minus Taylor Flats #1, were also listed in the later manuals and are considered long-term locations as discussed in Section 3.0, "Evaluation of Long-Term Sampling Locations."

Descriptions given in the locations manuals were essentially identical with the following two exceptions. According to the 1991 manual, the specific sampling site at the Wye Barricade was moved several hundred yards from the west side of Route 4S to the east side of Route 4S. The move apparently was made in the late-1980s when the road was relocated and the guard house was replaced. The assigned EMA number remained the same.

The literal name and EMA number for the Byers Landing sampling location have not changed since 1971. However, the location descriptions and photographs provided in the 1979 and 1983 locations manuals differed from those given in the 1991 locations manual. The earlier versions stated that the specific sampling site was "...located to the east, across the road from the air sampler." The 1991 version stated, "Continue east across the canal (from the air sampler) for 0.1 mile. Samples are collected from an undisturbed area on the left (north) side of the road." The location was changed because the original field was plowed and planted with alfalfa. According to a comment in the data base, the first sample was collected at this new location in June 1992.

Photographs of soil sampling locations shown in the 1979 locations manual were noted as being similar to those appearing in later versions except for the Wye Barricade and Byers Landing. Box #134924 retrieved from storage contained copies of the original photographs used in the 1979 locations manual. The historical photographs were used to verify the current soil sampling locations (see Section 3.0, "Evaluation of Soil Sampling Locations").

Many of the soil sampling locations were originally associated with "control plots." A PNNL monthly report prepared in 1972^(a) contained the following statement:

"The new Environmental Monitoring sites are now being posted. This will standardize the identification signs of all control plots, soil sample and vegetation plots and shoreline survey sites. Control plot signs only will be numbered."

These same identification signs are still present at onsite sampling locations and three perimeter offsite locations as noted in the locations manuals. The signs apparently were never placed on private land, and the three perimeter locations were associated with property originally under government control. The signs have a notation at the bottom "DUN Signs—1-72." Douglas United Nuclear, Inc. (DUN) apparently operated the sign shop at the Hanford Site in 1972.

2.2.3 Master Schedules and Annual Environmental Reports

Master schedules have been published in one form or another since environmental monitoring began at Hanford in the mid-1940s. Master schedules published annually in the early 1970s (and later) listed the sample types (including soil), locations, and corresponding EMA numbers of samples to be collected. Annual environmental reports^(b) have been published since 1957 to summarize and discuss results from environmental monitoring activities at Hanford. "Environmental Status Reports" emphasized data for locations on the Hanford Site (so-called onsite reports) and were published from 1965 through 1983. "Environmental Surveillance Reports" emphasized data for locations off of the Hanford Site (so-called offsite reports) and were published from 1957 to the present.

(a) *PNNL Monthly Report - January 1972*, M. W. Leale to J. P. Corley, January 25, 1972, archives of historical records.

(b) All annual environmental reports prepared for the Hanford Site to 1986 are listed in Price (1988).

A review of master schedules showed that EMA numbers and the general literal names of locations have remained the same over the years, with one exception. The "200 East Hill" sampling location apparently was established in 1971 at Control Plot (CP) #61, where a telephone line crossed Highway 4S near the bottom of the hill east of the 200 East Area^(a). However, the literal name given in the master schedule for calendar year 1978 was the "200 East Hill Air Sampling Station." This indicated that the soil sampling site was relocated to the top of the 200 East hill near the air sampling station and CP #15. However, the annual Environmental Status Reports for the Hanford Site reported data for "200 East Hill" in 1971 through 1973 and for "CP #61" in 1974 and 1975. Maps in the annual Environmental Surveillance Reports for 1971 through 1975 showed the general location of soil sampling to be near CP #61. In 1977, the literal "200 East Hill" was again used, and the map location appeared to be near CP #15 at the top of the 200 East hill (the 1976 report did not show data for all soil sampling locations). In 1983, the literal name was changed to "200 ESE" (same as the name change for the air sampling station), but the map location near CP #15 remained the same. Thus, the location for collecting soil samples from near the southeast corner of the 200 East Area was probably moved in 1976 or 1977 from the bottom of the 200 East hill to the top of the hill, about 2.25 miles closer to the Plutonium Uranium Extraction (PUREX) Plant stack than the original location.

The literal name and the EMA number of the Benton City sampling location have not changed since 1971; however, available records show that the specific sampling location may have been changed. A hand-drawn map accompanying a trip log recording the collection of special samples in 1970 showed a Benton City sampling location at the 614 Building air sampling station near the Public Utility District (PUD) electrical substation off Horn Road, north of Benton City proper. However, all locations manuals described the Benton City location as 4.8 miles south of Highway 240 on Horn Road. This location is near the old southern boundary of the Hanford Site and several miles north of Benton City. Annual reports from 1971 through 1982 showed map locations indicating the sampling location to be on the outskirts of Benton City. The 1983 report showed the map location as described in the 1979 and subsequent locations manuals. Thus, the Benton City soil sampling location may have been relocated several miles north of the original location, toward the Hanford Site. The year of change was not recorded, but four new offsite sampling locations were established in 1977, and "Benton City" was again sampled after two years of no sampling. Perhaps the sampling site for "Benton City" was changed in 1977 to the current location, and samples collected in 1971 through 1974 were collected near the PUD electrical substation.

2.2.4 Summary of Information on Sampling Locations

Literal names, EMA numbers, and locations for all long-term soil sampling sites listed in the locations manuals, master schedules, annual reports, and the results logbook appear to have remained the same since 1971 with the following exceptions. The literal names of several soil sampling locations have incurred minor name changes over the years. These a.k.a. names are noted in Section 3.0, "Evaluation of Soil Sampling Locations." Four sampling locations apparently were physically relocated. The Wye Barricade location was relocated across the road from the original site in the late-1980s. The Byers Landing location

(a) The location was shown on an environmental monitoring map dated May 1973. However, there is no telephone line crossing at the bottom of the 200 East hill at the present time.

was moved about 0.1 mile east of the original site in time for the 1992 sampling. The 200 ESE location was moved about 2.25 miles closer to the PUREX Plant stack in 1976 or 1977. The Benton City location may have been relocated several miles closer to the Hanford Site in the mid-1970s.

2.3 Sample Collection Procedures

The first recorded description of a procedure used to collect soil samples was described at a technical symposium in 1971 (Corley et al. 1971):

“Initial sampling was done with a flat-bottomed scoop approximately 18 by 12 inches. An attempt was made to take only the top one-half inch of soil. Subsequent sampling has been done with a closed-top sampler to minimize variation in sample depth. An ordinary cellulose tape container gives a neat, sharp-edged, reproducible cut in our desert soils, 9 cm in diameter by 1.6 cm deep, provided no large gravels are present.”

Soil sampling for the routine environmental monitoring program was also described in the Technical Procedures Document (PNNL 1977). The following description of soil sampling was noted in the section on Sampling Methods under the heading Soil and Vegetation:

“Surface soil samples are collected from selected areas of the Hanford Site and vicinity by using a ‘cookie cutter’ sampler approximately 5 cm in diameter and 25 cm in depth. Generally 5 spots are sampled within the area which have a minimum of vegetation.”

This record is believed to be inaccurate. Perhaps the 5 cm was really the radius and the 25 cm depth should have read 2.5 cm (i.e., 10 cm in diameter and 2.5 cm deep as noted below). The term “cookie cutter” was accurate, however, because even today the soil sampling tool is referred to as a “cookie cutter,” in obvious reference to the original cellulose tape container noted by Corley et al. in 1971.

2.3.1 Annual Environmental Surveillance Reports and Special Reports

The onsite annual environmental reports^(a) for 1971 (Bramson and Corley 1972) and 1972 (Bramson and Corley 1973) referred only to “...samples of the top two inches of soil...” However, the data tables showed that the top two inches consisted of two separate, one-inch-deep samples. The 1973 onsite report (Nees and Corley 1975) repeated the “...samples of the top two inches of soil...” statement, but the accompanying data table recorded only a single result for most locations. The logbook of handwritten results and the computer data base file recorded only one result for 1973 and listed the respective EMA

(a) The onsite annual environmental report published from 1971 through 1983 was entitled *Environmental Status of the Hanford Site for CY-19xx*. The offsite report was entitled *Environmental Surveillance at Hanford for CY-19xx*. The onsite report was discontinued in 1983, and all data were combined into a single report renamed *Environmental Monitoring at Hanford for 19xx*.

number associated with the 0-1 inch soil layer. Profile samples, to a depth of 12 inches, were also collected at three locations and reported in the 1973 annual report. In all likelihood, the statement about the top 2 inches of soil was a typographical error in the 1973 report because the wording was identical to the 1972 report. The 1974 report (Fix 1975), and reports for subsequent years, specifically stated that samples were collected from the top inch of surface soil. The annual report for 1974 was the first annual report to provide details on sampling:

“Each soil sample represents the composite of five ‘plugs’ of soil from an approximate 10 m² area. Each plug was approximately 2.5 centimeters (1 inch) in depth and 10 centimeters (4 inches) in diameter.”

Later, the sampling procedure was described in a special report reviewing results from the first 6 years of soil sampling for the routine environmental monitoring program (Miller et al. 1977):

“During the years 1971 through 1976, 136 soil and vegetation samples were collected from the Hanford environs. ...Each soil sample analyzed was a composite of 5 ‘plugs’ of soil collected from an area approximately 10 meters square. Each plug was about 2.5 centimeters in depth and 10 centimeters in diameter.”

2.3.2 Procedures Manuals

The first procedures manual may have been prepared in the mid- or late-1970s similar to the locations manual, but a copy was not located for this review. The earliest written procedure for soil sampling found in the historical record was a reference dated to 1984 (in Price et al. 1986). The selection of a specific sampling site was described as:

“Select five spots at least 10 paces apart within the desired sampling area which have a minimum of surface vegetation. If the sample site comes under cultivation or is otherwise disturbed, the EM supervisor should be notified so that an alternate site may be selected. Avoid sampling spots which are wind eroded, rocky, or disturbed significantly by animals.”

A PNNL memo written in 1971^(a) regarding the collection of mud, soil, and vegetation samples for pre-operational sampling at the construction site of the 331 Building stated:

“For soil samples, try to find undisturbed soil as close as possible to indicated spots (on attached map).”

Apparently, the objective has always been to collect soil samples from relatively undisturbed areas.

(a) PNNL Memo, J. P. Corley to W. C. Horton, *Pre-Operational Sampling - 331 Building*, February 16, 1971.

2.3.3 Summary of Information on Sample Collection Procedures

The historical records show that soil sampling for the routine environmental monitoring program has not changed significantly since 1971. Five plugs of soil, 2.5 cm deep and 10 cm in diameter, were collected at each sampling location within a relatively small area, placed into a common container (i.e., composited), labeled, and delivered to the analytical laboratory for radiochemical analysis. A minimum amount of vegetation was included with the soil sample. Since 1984, and probably since 1971, soil samples were to be collected from relatively undisturbed areas.

2.4 Analytical Procedures

One way to discover if analytical procedures have changed significantly since 1971 is to evaluate the usage of isotope identifying codes, i.e., numerical designators established by PNNL and originally used by U.S. Testing Co., Inc. to report analytical results for specific isotopes. The original 1971 version of the EMA Program Users Manual, Section V, "Isotope Codes," listed the following codes (in addition to others): 121 for strontium-90, 024 for cesium-137, 103 for plutonium-239 and 104 for uranium (Blummer and Corley 1971). A revised page in the manual dated July 1, 1974, indicated the addition of the isotope code 102 for plutonium-238. However, plutonium-238 analytical results were recorded in the logbook of handwritten results and the computer data base beginning in 1971. In a 1977 revision of the EMA Program User's Manual (Blummer and Fix 1977), Section V, "Isotope Codes," noted the following code changes: 100 for plutonium-239, 240 and 103 for plutonium. Thus, isotope code 103 in the historical record referred to both plutonium-239 and plutonium (or total plutonium) as noted below. The use of different isotope codes for plutonium was probably associated with the different analytical techniques used to detect plutonium in soil samples.

Although not well documented, it appears that some of the original plutonium data for soil samples analyzed by U.S. Testing Co., Inc. (e.g., as reported by Corley et al. 1971) were obtained by extracting and isolating the plutonium from the sample, electroplating it onto a stainless steel disc, and then exposing the disc to photographic film. The number of alpha tracks recorded on the film (produced by all isotopes of plutonium present) were counted by visual inspection with a microscope, converted to picocuries per unit of sample (pCi/g), and reported^(a). Such samples would have been reported under isotope code 103 for plutonium or plutonium-239. This was the same as the routine procedure used by U.S. Testing Co., Inc. for bioassay samples that were reported as total plutonium. Some of the early plutonium soil analyses (also reported in Corley et al. 1971) were analyzed in laboratories operated by PNNL and utilized a similar extracting and isolating technique. However, the resulting solution was evaporated onto a stainless steel disc and counted electronically with a silicon surface-barrier detector to determine the isotopes of

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- (a) Each alpha track was created by one disintegration of a plutonium atom. The number of disintegrations per minute (d/m) of film exposure was calculated and related to the weight (g) of the sample. The calculated d/m per gram was converted to picocuries per gram ($2.22 \text{ d/m/g} = 1 \text{ pCi/g}$) and reported as the result.

plutonium separately (so called "alpha spectrometry"). These results were reported under isotope codes 102 for plutonium-238 and 100 for plutonium-239,240. By 1972, U.S. Testing Co., Inc. was also analyzing certain samples by alpha spectrometry^(a). Counting uncertainties (error terms) consisting of \pm two standard deviations of the count rate were not routinely reported by U.S. Testing Co., Inc. until 1977. Beginning in 1984, both the counting uncertainty and \pm two times the total propagated uncertainty (for the entire analytical procedure) were reported by the analytical laboratory as a part of each result.

The annual environmental surveillance report for 1971 (Bramson and Corley 1972) recorded the analytical procedures used by U.S. Testing Co., Inc. at the time. Gamma scans to detect cesium-137 and other gamma-emitting radionuclides were performed with a lithium-drifted germanium (GeLi) detector. Strontium-90 and isotopes of plutonium were analyzed using chemical separation and specific analyses. Air and water samples were analyzed for total plutonium by the photographic film alpha track method. Solid foodstuffs, vegetation, and soil were analyzed for plutonium isotopes using alpha spectrometry. The film-track results were reported under isotope code 103 for (total) plutonium, and the alpha spectrometry results were reported under the isotope codes 102 for plutonium-238 and 100 for plutonium-239,240. Curiously, the PNNL computer data base shows results recorded in 1971 and 1972 (for both the 0-1 inch and 1-2 inch soil layers) for isotope codes 100 (plutonium-239,240) and 103 (plutonium). In most cases, the results for isotope code 100 are a three-decimal-place rounding of the results for isotope code 103. The sampling dates for each entry are the same. Beginning in 1973, the only isotope codes listed in the computer data base for routine soil samples are 100 for plutonium-239,240 and 102 for plutonium-238. The reason for the double entries is unknown, but it is improbable that the samples collected in 1971 and 1972 were analyzed twice. It is more likely that the addition of isotope code 103 to the data base required the transfer of data from one isotope code to another, and the original data were retained. Analytical results for routine soil samples reported in the 1971 annual report included plutonium-238 and plutonium (assumed to be plutonium-239,240). However, the samples must have been analyzed by alpha spectrometry because plutonium-238 could only be detected by that method.

2.4.1 Summary of Information on Analytical Procedures

The isotope identifying codes used to report strontium-90 or cesium-137 have not changed since 1971. However, plutonium data for soil samples analyzed before 1972 by U.S. Testing Co., Inc. were probably the result of a photographic film-alpha track procedure and were understood to represent total plutonium. Results were reported under isotope code 103. Soil samples analyzed in 1972 (i.e., samples collected in both 1971 and 1972) and in later years were analyzed by alpha spectrometry, i.e., a silicon surface-barrier detector system capable of separately identifying plutonium-238 and plutonium-239,240 (the technique, as today, did not differentiate between the isotopes plutonium-239 and plutonium-240). Results were reported under isotope code 100. The annual environmental surveillance report for 1971 (Bramson and Corley 1972) recorded the analytical procedures used by U.S. Testing Co., Inc. at the time and stated that

(a) The first routine soil samples collected for the environmental monitoring program (i.e., samples not collected for a special purpose) were obtained in September and October 1971, but apparently were not submitted to U.S. Testing Co., Inc. for analysis until April 1972.

soil samples were analyzed by alpha spectrometry. Thus, all results from the analyses of routine soil samples reported in annual reports since 1971 were apparently determined by alpha spectrometry and included plutonium-238 and plutonium-239,240.

2.5 Previously Unreported Results

An attempt was made in 1987 to review the analytical results for all historical soil samples collected for the routine environmental monitoring program (Price 1988). Some new information was discovered during the current review. Before 1970, the few special soil samples collected were usually analyzed only for total (gross) beta, total (gross) alpha, strontium (89 and 90 isotopes), and a gamma scan was run, primarily for ruthenium-106 and cesium-137.

A spread of plutonium contamination to the offsite environment at the U.S. Atomic Energy Commission's (AEC) Rocky Flats Plant was discovered in the late 1960s, which led to concern and speculation about contamination in the environs at all AEC facilities that handled plutonium. An AEC letter^(a) from Washington D.C. Headquarters to the Richland and Savannah River Operations Offices in late 1969 noted the situation at Rocky Flats and asked "...do you sample and measure the plutonium content of soil at your site...?" The answer was "no," and the inquiry provided the initiative to collect soil samples from the Hanford Site and vicinity for plutonium analyses. Soil samples were first collected in February 1970 from both onsite and offsite locations. Additional samples were collected later in 1970 and in 1971. Results were reported by Corley et al. (1971) at a scientific symposium held in Los Alamos, New Mexico. Soil samples were collected on a routine basis beginning in 1971, and results have been published in annual environmental reports from 1971 through 1994. Since 1989, the need for collecting additional soil samples has been evaluated each year.

Results for samples not previously reported in annual or special reports for the years 1970 through 1972 are listed in Appendix A. No other records of unreported results were located in the historical files during this review.

2.5.1 Summary of Unreported Results Listed in Appendix A

Unreported results were found in historical records for the locations listed below. None of the results was noted to be extraordinary.

1970

1. Two samples were collected from an area northeast of the Jersey Nuclear uranium fuel manufacturing plant (currently Siemens Power Corp.). Samples were analyzed by U.S. Testing Co., Inc.

(a) AEC Letter, from F. P. Baranowski (Washington D.C.) to D. G. Williams (Richland Operations Office), *Soil Contamination by Reactor Products*, December 19, 1969.

2. Four offsite samples from the general area of Byers Landing were collected jointly with the AEC Division of Compliance. A composite background sample was collected also. Analyses were performed by an AEC laboratory at the Idaho National Engineering Laboratory, Idaho Falls, Idaho.

1971

1. Ten samples were collected from four locations at the construction site of the Fast Flux Test Facility (FFTF) as part of a pre-operational survey. Samples were analyzed by U.S. Testing Co., Inc.
2. Five samples were collected as part of a pre-operational survey at the 331 Building. Samples were analyzed by U.S. Testing Co., Inc.
3. A composite of four samples collected from the Wahluke Slope across the River from the 100-K Area and 100-N Area was analyzed by U.S. Testing Co., Inc.
4. Five soil samples, from three offsite and two onsite locations, were analyzed by U.S. Testing Co., Inc.

1972

1. Eleven samples were collected from five offsite locations, five onsite locations, and Locke Island. Samples were analyzed by U.S. Testing Co., Inc.
2. Three soil samples and one mud sample were collected from the vicinity of Honey Hill Pond (a.k.a. West Lake). Samples were analyzed by U.S. Testing Co., Inc.
3. Samples collected from three onsite locations near 200 East Area were analyzed by U.S. Testing Co., Inc.

2.6 Summary of Reported Results

Results from the analyses of soil samples collected routinely each year were reported in annual reports. In addition, three reviews and summaries have been prepared over the years (Miller et al. 1977, Price 1988, Poston et al. 1995). However, for completeness, Appendix B lists data for soil samples collected at the 20 long-term sampling locations and two supplemental locations. Supplemental locations in proximity to other long-term sampling sites are included to enhance the long-term record. The results shown for each sampling location represent all data in the PNNL computer data base for strontium-90, cesium-137, plutonium, plutonium-238, and plutonium-239,240 for 1971 through 1994 (the last year of sampling).

The supplemental locations are "FFTF Control Plot 62" and "ERC." The FFTF Control Plot 62 supplements both the NE FFTF and SE FFTF sampling locations. The FFTF Control Plot 62 was sampled from 1971 through 1974 and was located between the other FFTF locations. The NE FFTF and SE FFTF locations were sampled from 1975 through 1994. ERC is located several hundred yards from the ALE sampling site. ERC was sampled from 1971 through 1974; ALE was sampled from 1975 through 1993.

2.7 Conclusions for the Review of Historical Records

Environmental monitoring program historical files and annual reports were reviewed for information and documentation on the following issues:

- **Have soil sampling locations changed significantly over the years?**

Literal names, EMA numbers, and locations for all soil sampling sites listed in the locations manuals, master schedules, annual reports, and the results logbook appear to have remained the same since 1971, or the year the location was established, with the following exceptions. The literal names of several soil sampling locations have incurred minor name changes over the years. These a.k.a. names are noted in Section 3.0, "Evaluation of Soil Sampling Locations." Four sampling locations apparently were physically relocated. The Wye Barricade location was relocated across the road from the original site in the late-1980s. The Byers Landing location was moved about 0.1 mile east of the original site in 1992. The 200 ESE location was moved about 2.25 miles closer to the PUREX Plant stack in 1976 or 1977. The Benton City location was relocated several miles closer to the Hanford Site in the mid-1970s.

- **Have soil sampling procedures changed significantly over the years?**

Soil sampling procedures have not changed significantly since 1971. Five plugs of soil, 2.5 cm deep and 10 cm in diameter, were collected at each sampling location, placed into a common container (i.e., composited), labeled, and delivered to the analytical laboratory for radiochemical analysis. A minimum amount of vegetation was included with the soil sample. Records show that since 1984, and probably since 1971, soil samples have been collected from relatively undisturbed fields.

- **Have significant changes occurred over the years in analytical procedures used to detect plutonium in soil samples or to report data?**

The isotope code identifiers used to report strontium-90, cesium-137, or uranium have not changed since 1971. However, plutonium data for soil samples analyzed before 1972 by U.S. Testing Co., Inc. were analyzed by a photographic film-track method and were understood to represent total plutonium. Results were reported under isotope code 103. Routine soil samples analyzed in 1972 (i.e., samples collected in both 1971 and 1972) and later years were processed using a silicon surface-barrier detector system capable of separately identifying the isotopes plutonium-238 and plutonium-239,240 (the technique, even as used today, does not differentiate between the isotopes plutonium-239 and plutonium-240). Results were reported under isotope code 100. All results from the analyses of routine soil samples reported in the annual reports since 1971 were determined by alpha spectrometry and included the isotopes of plutonium-238 and plutonium-239,240.

- **Have all significant results for plutonium and other important long-lived radionuclides detected in soil samples been reported?**

Previously unreported results from soil sampling for 1970, 1971, and 1972 were located in archived historical records and are listed in Appendix A. None of the results are considered extraordinary.