Y/TS-1704

Y-12

OAK RIDGE
Y-12
PLANT

LOCKHEED MARTIN

RADIOLOGICAL MONITORING PLAN

FOR

THE OAK RIDGE Y-12 PLANT: SURFACE WATER

October 1997

Water Compliance Department

Environmental Compliance Organization

Oak Ridge Y-12 Plant
Oak Ridge, Tennessee 37831

Managed by
Lockheed Martin Energy Systems, Inc.
for the
U.S. Department of Energy
under Contract DE-AC05-84OR21400

MANAGED BY
LOCKHEED MARTIN ENERGY SYSTEMS, INC.
FOR THE UNITED STATES
DEPARTMENT OF ENERGY

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TABLES

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ACRONYMS

ALARA	As low as reasonably achievable
ANPR	Advance Notice of Proposed Rulemaking
ANSI	American National Standards Institute
AS	Alpha Spectrometry
ASER	Annual Site Environmental Report
BC	Bear Creek
BCK	Bear Creek Kilometer
CWA	Clean Water Act
DCF	Dose conversion factor
DCG	Derived concentration guide
DL	Detection limit
DMR	Discharge Monitoring Report
DOE	U.S. Department of Energy
EFPC	East Fork Poplar Creek
ESLIMS	Energy Systems Laboratory Information Management System
EMMIS	Environmental Monitoring Management Information System
EPA	U.S. Environmental Protection Agency
ERPP	Environmental Radiation Protection Plan
MCL	Maximum contaminant level
MDA	Minimum detectable activity
MS	Mass Spectrometry
NPDES	National Pollutant Discharge Elimination System
ORNL	Oak Ridge National Laboratory
ORR	Oak Ridge Reservation
PQL	Practical quantitation level
QA	Quality assurance
RMP	Radiological Monitoring Plan
SS	Settleable solids
TDEC	Tennessee Department of Environment and Conservation
TEDE	Total effective dose equivalent
TSS	Total suspended solids

1. INTRODUCTION AND BACKGROUND

1.1 PURPOSE

The Oak Ridge, Tennessee, Y-12 Plant is operated by Lockheed Martin Energy Systems, Inc., (Energy Systems) for the United States Department of Energy (DOE). The plant operates within the framework of DOE Orders and, more recently, local, state, and federal regulations. The Y-12 Plant conducts a surface water monitoring program in response to DOE Orders and state of Tennessee requirements under the National Pollutant Discharge Elimination System (NPDES). The anticipated codification of DOE Order 5400.5 for radiation protection of the public and the environment (10 CFR Part 834) will require an environmental radiation protection plan (ERPP). The NPDES permit issued by the state of Tennessee requires a radiological monitoring plan (RMP) for Y-12 Plant surface waters.

In a May 4, 1995, memo, the state of Tennessee, Division of Water Pollution Control, stated their desired needs and goals regarding the content of RMPs, associated documentation, and data resulting from the RMPs required under the NPDES permitting system (L. Bunting, General Discussion, Radiological Monitoring Plans, Tennessee Division of Water Pollution Control, May 4, 1995). Appendix A provides an overview of how the Y-12 Plant will begin to address these needs and goals. It provides a more complete, documented basis for the current Y-12 Plant surface water monitoring program and is intended to supplement documentation provided in the Annual Site Environmental Reports (ASERs), NPDES reports, Groundwater Quality Assessment Reports, and studies conducted under the Y-12 Plant Environmental Restoration (ER) Program.

The purpose of this update to the Y-12 Plant RMP is to satisfy the requirements of the current NPDES permit, DOE Order 5400.5, and 10 CFR Part 834, as currently proposed, by defining the radiological monitoring plan for surface water for the Y-12 Plant. This plan includes initial storm water monitoring and data analysis. Related activities such as sanitary sewer and sediment monitoring are also summarized. The plan discusses monitoring goals necessary to determine background concentrations of radionuclides, to quantify releases, determine trends, satisfy regulatory requirements, support consequence assessments, and meet requirements that releases be "as low as reasonably achievable" (ALARA).

The RMP has evolved from a plan that meets only NPDES requirements to one that considers other monitoring goals to meet the needs of other users to the extent practical. Although the monitoring goals for other users are not requirements of the NPDES RMP, to the extent that they can be accommodated at reasonable costs, lower long-term costs may result.

These aspects of the plan are included, not for formal approval by the Tennessee Department of Environment and Conservation (TDEC), Division of Water Pollution Control, staff, but to address their desired needs and goals and to ensure that a cohesive and complete discussion of the subject has been given. Regulatory authority for requirements of DOE Orders rests with DOE.

1.2 BACKGROUND

Current guidance and regulatory documents as well as the roles of agencies involved in regulating and monitoring the Y-12 Plant water monitoring program are summarized below. The state of Tennessee Division of Water Pollution Control monitoring goals and requirements are summarized in Appendix A. In addition, surface water monitoring is part of the responsibility of the DOE Oversight Division of the TDEC.

The most current DOE guidance is contained in the January 1991 report *Environmental Regulatory Guide for Radiological Effluent Monitoring and Environmental Surveillance* (DOE/EH-01T3). This regulatory guide presents elements of a radiological effluent monitoring and environmental surveillance program considered acceptable to DOE in support of DOE Order 5400.5 (*Radiation Protection of the Public and the Environment*) and DOE Order 5400.1 (*General Environmental Protection Program*).

DOE has proposed to codify the requirements of DOE Order 5400.5 into the Code of Federal Regulations as 10 CFR 834 (Part 834). The codification will strengthen the Order by establishing enforcement procedures and penalties for noncompliance. The schedule for final promulgation of the rule is uncertain at this time.

Essentially all of the requirements in Part 834 are required in DOE Order 5400.5. Since the current NPDES Y-12 Plant RMP considers the requirements of DOE Order 5400.5, updates will focus on any new details and/or changes provided under Part 834. Since Part 834 has not been issued in final form at this writing, future modifications will be covered through revisions of this plan. In order to fully satisfy the requirements of Part 834, this plan must be combined with plans for other media at the Y-12 Plant and with other plans for all media at other local sites.

Currently, the DOE regulatory guide identifies high priority items with the word *should* followed by an asterisk (should*). Guidance items are denoted using only the word *should.* High priorities pertinent to all monitoring data include:

- "The required detection levels of the monitoring and analysis systems should* be sufficient to demonstrate compliance with all regulatory requirements. . . ."
- "Sampling systems should* be sufficient to collect representative samples that provide for an adequate record of releases, to predict trends, and to satisfy needs to quantify releases. . . ."
- "Specific analytical methods should* be identified, documented and used to identify and

quantify all radionuclides in the facility inventory or effluent that contribute 10% or more to the public dose or environmental contamination associated with the site. . . ."

- "The statistical techniques used to support the concentration estimates, to determine their corresponding measures of reliability, and to compare radionuclide data between sampling and/or measurement points and times should* be designed with consideration of the characteristics of effluent and environmental data."

The NPDES permit generally requires use of U.S. Environmental Protection Agency (EPA) methods or other methods approved by the state. There are no EPA regulations for non-drinking water supplies. The most pertinent general federal regulatory requirements with respect to drinking water monitoring are contained in the Safe Drinking Water Act (40 CFR 141.25) as follows:

"For the purpose of monitoring radioactivity in drinking water, the required sensitivity of the radioanalysis is defined in terms of a detection limit. The detection limit shall be that concentration which can be counted with a precision of plus or minus 100 percent at the 95 percent confidence level (1.95 σ where σ is the standard deviation of the net counting rate of the sample). . . ." Note that the Y-12 Plant effluents and surface water streams are not drinking water supplies and are not subject to the requirements of the Safe Drinking Water Act. Provisions of the Act are used here as reference values for perspective on surface water monitoring.

The most recent EPA-proposed regulations on radionuclides in drinking water are contained in an Advanced Notice of Proposed Rulemaking (ANPR) *National Primary Drinking Water Regulation; Radionuclides* (40 CFR 141,142, *Federal Register* 56, No. 138, July 18, 1991).

The ANPR presents a selection of analytical methods based on accuracy of recovery (lack of bias) and precision (good reproducibility over the range of MCLs considered). The EPA concluded (Table 15, p. 33092; see Table 6) that analytical methods with acceptable precision/accuracy over a range of concentrations, including the MCLs, are technically and economically available for radionuclide monitoring of processed drinking water supplies. The MCLs are included in this RMP only as reference concentrations. Drinking water concentrations do not apply to NPDES outfalls. Generally, outfall concentrations that approach allowable drinking water concentrations would lead to negligible contributions to actual drinking water.

Taken together, the DOE high priorities and the EPA regulations lead to the following data quality objectives:

- The monitoring goals for detection limits (DLs) for radiological contaminants must be less than the standards with which they are compared.
- Statistical precision of the DLs must be plus or minus 100 percent at the 95 percent confidence level or better.

The general requirements of the Y-12 Plant NPDES Permit reinforce the DOE Regulatory Guide and Clean Water Act for surface water. The specific objectives of the Radiological Monitoring Plan are as follows:

(1) Develop a routine radiological monitoring program to demonstrate compliance with Part III, H of the Y-12 Plant NPDES Permit;

(2) Document plans for ensuring that discharges of radioactive materials are ALARA to meet DOE requirements; and

(3) Describe methods for identifying and locating sources of radioactive discharges, if any, other than those in NPDES monitoring locations (outfalls).

To demonstrate the ALARA philosophy, it is useful to have goals (e.g., concentrations), with which measured or estimated concentrations can be compared for each radionuclide. Regulations have not been developed for each radioisotope of possible concern at the Y-12 Plant; therefore, monitoring goals have not been clearly identified. However, DOE has established derived concentration guides (DCGs) equivalent to 100 mrem/yr by each pathway for all radioisotopes; the state calls for comparison of measured concentrations with the DCGs.

DOE Order 5400.5 requires that operations be conducted to ensure that doses to members of the public and releases of radioactive material into the environment:

- do not exceed established dose limits or DOE release limits, and
- are kept ALARA.

The primary public dose limit is 100 mrem/yr total effective dose equivalent (TEDE) to individuals in the public from all radiation sources and exposure pathways combined. Doses from background radiation or diagnostic or therapeutic medical procedures are not included.

DOE recognizes a need for a threshold level for source identification because of the inherent burden of identifying all other man-made radiation sources in the community or area.

If the projected or estimated dose to any members of the public from a DOE facility is:

- less than 30 percent of the 100 mrem/yr limit (< 30 mrem/yr), then the dose contributors from other non-DOE sources need not be assessed;
- greater than 30 mrem/yr, then the dose contribution of other non-DOE sources have to be assessed; and

- if the contribution of any other non-DOE source is greater or equal to 30 mrem/yr, then it and the dose from the DOE activity shall be added in order to determine compliance with the 100 mrem/yr limit.

DOE Order 5400.5 has a requirement to report to DOE if the dose to any member of the public exceeds 10 mrem/yr. This limit is taken to be numerically equivalent to the 10-mrem/yr limit applied separately to airborne and liquid effluents. The overall implication of this limit is that DLs must be based on a dose limit of less than 10 mrem/yr to meet the reporting requirements. Monitoring goals for each radioisotope must be based on a lower dose limit since the integrated dose from all isotopes determines compliance with the 100-mrem/yr limit.

The NPDES Permit (Part III, H) requires that data for isotopes must be at least as precise as is necessary to evaluate results relative to DCGs in DOE Order 5400.5. Therefore, NPDES monitoring results are compared with DCGs. Detection limits or minimum detectable activities (MDAs) based on monitoring goals for various uses are discussed in Section 3.

1.3 PLAN ORGANIZATION

The Radiological Monitoring Plan is organized as follows.

Section 2.	Y-12 Plant Monitoring Program
Section 3.	Detection Limit Monitoring Goals
Section 4.	Monitoring Methodologies to Meet EPA and Tennessee Requirements
Section 5.	Data Management and Radiological Analysis
Section 6.	Quality Assurance
Appendix A:	Overview Radiological Analysis of the Y-12 Plant Surface Water Radiological Monitoring Program

2. Y-12 PLANT MONITORING PROGRAM

A radiological monitoring program is in place to address compliance with DOE Orders, the NPDES permit, and the ALARA philosophy. Monitoring is performed routinely and results will be reported each quarter to the state of Tennessee as an attachment to the monthly Discharge Monitoring Report (DMR) for NPDES compliance. Additional monitoring associated with various source identification projects or remedial investigations conducted as part of environmental restoration activities at the Y-12 Plant are not performed as part of routine NPDES compliance monitoring and therefore will not be submitted with the DMR. Data obtained from supplemental programs will be available in reports issued as part of the individual programs.

In the Y-12 Plant monitoring plan, surface water is monitored at points that reflect individual facilities as well as at points that reflect the combined contributions of all facilities. DOE Order 5400.5 provides that dose estimates consider contributions from all facilities and are thus reflected in the monitoring plan for Y-12 Plant surface water. Determining TEDE to the public is appropriately based on the monitoring points that include the contribution of all facilities at the Y-12 Plant. This monitoring plan does not consider other potential routes (i.e., airborne releases and food chains). Thus, a complete determination of TEDE cannot be made based on this plan alone. The other routes from the Y-12 Plant and all routes from other DOE facilities on the Oak Ridge Reservation (ORR) (e.g., Oak Ridge National Laboratory [ORNL], East Tennessee Technology Park) must be considered in order to satisfy the order requirements. Determination of TEDE from all sites and pathways is done through the use of dose-assessment models and is documented in the ASER. This monitoring plan provides adequate monitoring goals for Y-12 Plant surface water releases to provide input of sufficient sensitivity and accuracy to reliably determine the Y-12 Plant surface water component of the TEDE.

The routine radiological monitoring program is designed to monitor effluents at three types of locations: (1) treatment facilities, (2) other point and area source discharges, and (3) instream locations. With this sampling and analysis program, data will be obtained on primary point sources as well as on locations that represent the composite of other potential sources. This plan will be reviewed periodically to determine necessary modifications to the sampling frequencies, parameters, and locations. Modifications, if any, will be based on the analysis of the previous year's data and its effectiveness in satisfying the objectives of this plan.

Tennessee requirements are specified in the permit—including sampling location, frequency, sample type, and parameters. Appendix A documents the basis of the parameters monitored based on operational history, expected chemical and physical relationships, and historical monitoring results.

2.1 TREATMENT FACILITIES

The facilities to be monitored include outfalls:

501	Central Pollution Control Facility
502	West End Treatment Facility
503	Steam Plant Wastewater Treatment Facility
512	Groundwater Treatment Facility
520	Lithium Process Condensate
551	Central Mercury Treatment Facility

Most process wastewaters are collected and treated at one of these facilities. Treated process wastewaters, other wastewaters, ground water, cooling waters, cooling tower blowdown and storm waters may be released through Outfall 200. These process wastewaters may contain radioactive constituents. Therefore, the points of discharge from these facilities are ideal locations for monitoring radioactivity levels before the discharges enter the receiving water.

2.2 OTHER POINT AND AREA SOURCE DISCHARGES

The discharges to be monitored include the following outfalls:

S17	Kerr Hollow Quarry
S19	Rogers Quarry

These outfalls could contribute to movement of radionuclides off site in water that is not treated at one of the treatment facilities. The Isotope Separation Process, formerly monitored at Outfall 142, primarily separated stable isotopes using calutrons. However, one calutron was reserved for processing small amounts of uranium isotopes (^{233}U, ^{235}U, ^{238}U) and small amounts of some actinides (curium isotopes and ^{242}Pu). Low or no flow at Outfall 142 and using the process only for separating stable isotopes in recent years makes monitoring for radioisotopes unnecessary.

2.3 INSTREAM MONITORING POINTS

The instream locations to be monitored include:

Station 304	Bear Creek kilometer 4.55 at Highway 95
9422-1	Station 17 on East Fork Poplar Creek
200	North/South Pipes on East Fork Poplar Creek

In addition to monitoring specific point and area source discharges, it is necessary to monitor several instream locations to determine ambient or composite radioactive constituents. Stations 17 and 304 represent the water crossing the Y-12 Plant boundary and moving offsite. These monitoring sites will be used to demonstrate instream compliance and to provide data for mass balance evaluation of radioactive inputs. Surface water monitoring at Bear Creek kilometer 11.97 has been conducted as part of the Groundwater Monitoring Program. The Y-12 Groundwater Monitoring Program monitors this location semi-annually. Surface water monitoring requirements of this RMP will be added to the groundwater program. Monitoring at Station 8 on East Fork Poplar Creek was discontinued, but station 200 was added to meet requirements of the NPDES permit. Station 200 is upstream of Station 8 and represents the primary release point of the Y-12 Plant to East Fork Poplar Creek.

2.4 SAMPLING

Previous data indicate that 7-day composites must be collected at several locations to accumulate detectable quantities of radionuclides of concern. Detection limits or MDAs for radionuclides, as related to sample volumes and counting times are discussed in Section 3 and Appendix A, Section 3. General surface water sampling will also be undertaken at the Y-12 Plant under the NPDES program. Sample preservation will follow protocols required by the Lockheed Martin energy Systems (Energy Systems) Environmental Surveillance Procedures Quality Control Program. These procedures have been approved for use by the state and by EPA. EPA has identified the analytical methods as technically and economically available for radionuclide monitoring. The NPDES Permit, under Part I.B.2, requires that these methods be used for monitoring. The state has previously approved the Y-12 Plant specific procedures that are equivalent to the EPA standard methods listed in Table 6 of Section 4. They have acceptable DLs (± 100 percent at the 95 percent confidence level) and provide the techniques to follow trends to levels well below DOE and NPDES requirements.

Sample frequency, sample type, and parameters monitored at the Y-12 Plant outfalls and stations are given in Tables 1 through 3.

Table 1. The NPDES Radiological Monitoring Program at the Y-12 Plant Wastewater Treatment Facilities

Outfall	Facility	Sampling Frequency	Sample Type	Parameters
501	Central Pollution Control Facility	1/week	Composite[a]	Isotopic analysis[b]
502	West End Treatment Facility	1/week	Composite	Isotopic analysis[b]
503	Steam Plant Wastewater Treatment Facility	1/week	Composite	Isotopic analysis[b]
512	Groundwater Treatment Facility	1/week	Composite	Isotopic analysis[b]
520	Lithium Process Condensate	1/month	Grab	Isotopic analysis[b]
551	Central Mercury Treatment Facility	1/month	Composite	Isotopic analysis[b]

[a] Composite will be taken during batch discharge. If facility discharges continuously, a 24-hour composite will be taken.

[b] Specific isotopes for NPDES monitoring are listed in Section 2.5.

Table 2. The NPDES Radiological Monitoring Program at the Y-12 Plant for Point and Area Source Discharges

Outfall	Facility/Area	Sampling Frequency	Sample Type	Parameters
S17	Kerr Hollow Quarry	1/month	24-hour Composite	Isotopic analysis[a]
S19	Rogers Quarry	1/month	24-hour Composite	Isotopic analysis[a]

[a] Specific isotopes for NPDES monitoring are listed in Section 2.5.

Table 3. The NPDES Radiological Monitoring Program at the Y-12 Plant for Instream Locations

Monitoring Station	Location	Sampling Frequency	Sample Type	Parameters
304	Bear Creek	1/month	7-day Composite	Isotopic analysis[a]
17 9422-1	East Fork Poplar Creek	1/week	7-day Composite	Isotopic analysis[a]
200	North/South Pipes	1/week	24-hour Composite	Isotopic analysis[a]

[a] Specific isotopes for NPDES monitoring are listed in Section 2.5.

The NPDES permit (Part I) requires measurement frequencies of one per month and monthly composite sample types. The definition of a monthly composite sample for the purposes of the permit "is a sample made by combining at least 4 separate 24-hour composite samples collected in separate weeks of the months at a rate proportional to the flow. Individual composites may be analyzed separately and the results averaged and reported on the monthly composite sample. If sampling cannot be made for 4 separate weeks of the month, the monthly composite samples shall be composited for those week(s) of the month in which sampling could be made."

The average of four weekly measurements is considered equivalent to measurements made on monthly samples composited from weekly samples.

The measurement frequency in this plan corresponds to the sampling frequencies of one per week or one per month. Thus the measurements meet the requirements of the permit.

The permit (Part I.C.2.) indicates that the results of more frequent monitoring than described in the permit shall be indicated and included in the calculations and reported in the DMR or computerized data submission.

The NPDES permit requires (Part I) gamma scans in addition to isotopic analysis for all the locations listed in Tables 1, 2, and 3. Although operational history and previous monitoring results indicate that gamma emitters in Y-12 Plant surface water are below drinking water criteria, the permit requires that gamma scans be conducted. The NPDES permit (Part III.H) requires:

> “Sufficient data collection to allow determination of the appropriate parameters to be analyzed and reported for the radiological monitoring program. Data must be at least as precise as is necessary to evaluate results relative to the Derived Concentration Guide (DOE Order 5400.5).
>
> Where reasonable, the Division may require greater analytical precision. The permittee shall present a report summarizing the data to the Division of Water Pollution Control. If requested, the permittee shall make records of raw data available to the Division of Water Pollution Control. Initial data collection for re-evaluation of the radiological monitoring plan shall be completed and reported to the Division by August 1, 1995. Once assessment and reevaluation of the radiological monitoring plan is made, the plan may be modified to reflect appropriate monitoring.”

The results of the initial gamma scan were reported to the Division of Water Pollution Control. The results confirm previous operational history and monitoring results and gamma scanning may be eliminated from the RMP. (See Sections 2.5 and 5.2 for further discussion.) Gamma scanning is, therefore, not included in this version of the RMP. ^{90}Sr, ^{137}Cs, ^{3}H and ^{99}Tc will continue to be monitored as tracers for beta and gamma radionuclides although their concentrations in surface water are very low.

Their presence in groundwater and their mobility merits continued monitoring. (See Appendix A.)

Measurements of gross alpha, beta, or gamma can be useful as screening measurements when specific isotopes make a significant contribution to the total count, which is greater than the highly variable background total counts.

Based on a review of the data, gross measurements are not predictive of specific alpha-, beta-, or gamma-emitting isotopes for the low levels encountered in Y-12 Plant surface water. Gross measurements can be misleading since isotopic composition can vary widely for a given gross count. Limits for individual isotopes could be exceeded for a given gross limit.

Measurements of specific isotopes are required by DOE Order 5400.5 and by the state of Tennessee. Specific isotopes measured are based on analyses such as presented in Section 2.5 and Appendix A.

2.5 RADIOLOGICAL PARAMETERS MONITORED

The uranium isotopes to be monitored based on the analysis in Appendix A are listed below:

^{238}U,

^{235}U,

^{234}U, and

total U and weight percent ^{235}U.

These parameters reflect the major activity, uranium processing, throughout the entire history of the Y-12 Plant. Uranium concentrations are far higher than all other isotope concentrations, based on monitoring results through 1995. The isotopes ^{234}Th and ^{234}Pa$_m$ will be in secular equilibrium with ^{238}U unless there are processes in operation that enrich them relative to ^{238}U with direct release to surface water.

Fission and activation products to be monitored include:

^{90}Sr,

^{3}H,

^{99}Tc, and

^{137}Cs.

These parameters reflect a relatively minor activity at the Y-12 Plant— processing of recycled uranium from reactor fuel elements from the early 1960s to April 1988. A conservative analysis of expected relative isotopic concentrations assuming direct processing of fuel elements indicates ^{90}Sr concentrations potentially higher than other fission products. The isotope ^{3}H is not expected to be high in fuel elements—it is produced primarily as an activation product in reactor coolants. But ^{3}H is highly mobile and is detected in groundwater samples associated with the S-3 Pond Site. The isotope ^{99}Tc is also expected to be at relatively low concentrations as a fission product in reactor fuel elements. However, ^{99}Tc is also highly mobile and has been detected in groundwater samples associated with the S-3 Pond Site and at low levels in surface water. The isotopes ^{99}Tc and ^{3}H likely were brought to the Y-12 Plant in wastes from other sites. The Y-12 Plant received some materials from two other DOE facilities—the Savannah River Plant and the Idaho Chemical Processing Plant. In addition, wastes from the K-25 Site and ORNL have come to the Y-12 Plant. Based on expected relative concentrations and potential hazards discussed in Appendix A, other fission product concentrations should be very low and less hazardous than ^{90}Sr. Gamma spectrometry was performed to detect any significant concentrations of gamma emitters that may have been missed in this analysis. None were detected.

The conservative analysis given in Appendix A of the RMP which assumed direct processing of reactor fuel elements probably encompassed the majority of possibilities. However, the NPDES Permit required general gamma scans to investigate whether any detectable concentrations of gamma emitters had been missed in the analysis. Wastes from other sites were of particular interest because they might have come from other processes such as accelerator produced isotopes used in biological research or from the thorium fuel cycle. However, most radioisotopes used in biological research are low energy beta emitters. Earlier monitoring for radioisotopes used in biological research by ORNL at the Y-12 Plant yielded very low concentrations. See Section 5.2 for further analysis.

Transuranium isotopes to be monitored include the following:

^{241}Am,
^{237}Np (^{233}Pa),
^{238}Pu, and
^{239}Pu.

These isotopes are also related to recycle uranium processing. Concentrations were estimated to be low in fuel elements compared with ^{90}Sr, but toxicities are much higher. They are expected to be less mobile than ^{90}Sr. Concentrations have been low in surface water; but they are monitored because of their long half-lives, presence in groundwater, and long-term risk potential.

A comprehensive review of transuranium elements in groundwater, surface water, sewer and stormwater should be conducted to determine need for further monitoring. Additional information may be found in Section 5.2.

Isotopes associated with thorium processing and other natural radionuclides include:

^{232}Th,
^{230}Th,
^{228}Th,
^{226}Ra, and
^{228}Ra.

These isotopes are monitored to reflect previous thorium processing and to characterize important background radioisotopes. These isotopes could also interfere with analysis for contaminants (see Appendix A). Monitoring of these isotopes is performed to meet DOE Order requirements.

2.6 STORM WATER MONITORING

The NPDES permit also requires (Part III.H.) that the RMP be expanded to include storm water monitoring. A Storm Water Pollution Prevention Plan containing a Storm Water Monitoring Plan for the Y-12 Plant has been prepared. About 31 Category I, 33 Category II, 12 category III, and 4 storm water outfalls of special interest were identified in the storm water monitoring plan. Characterization of a single outfall will be made where several outfalls are very similar rather than characterizing each outfall in a group of similar outfalls. The permit requires a minimum of 25 storm water characterizations per year and requires certain outfalls in the first year. Outfalls are grouped into similar categories based on land use for the drainage area and possible pollutants. Category I outfalls are to be sampled twice during the permit period. Category II and III outfalls will be sampled once per year. Samples must be either flow weighted or time weighted.

During each sampling period associated with a storm event, a different site in each grouping will be sampled. If a grouping contains only one site, then that site will be sampled during every sampling period. If all sites in a multiple site grouping have been sampled, sampling on one site per grouping will continue throughout the life of the storm water plan.

In the Storm Water Monitoring Plan, each sample will be analyzed for the same isotopes as for surface water monitoring given in Section 2.5. Also, gamma scans using a high-resolution gamma spectroscopy system have been conducted. Initial data have been reported, as acquired, to the TDEC as an attachment to the DMR.

As is the case for general surface water monitoring at the Y-12 Plant site, uranium isotopes dominate relative to DCGs. The initial storm water monitoring data also confirm the results for general surface water monitoring—the analysis in Appendix A appears to be correct. That is, monitoring for ^{137}Cs among the gamma emitters and ^{90}Sr, ^{3}H, and ^{99}Tc among the beta emitters is adequate to monitor movement of fission and activation products in the storm water system. See Section 5.2 for further analysis.

3. DETECTION LIMITS AND MONITORING GOALS

Detection limits or minimum detectable activities (MDAs) vary according to the purpose of the activity. Detection limits required to meet monitoring goals based on NPDES permit requirements, DOE Order 5400.5, and preliminary remediation goals are discussed in this section. Greater detail may be found in Section 3 of Appendix A.

3.1 CONCENTRATION LIMITS FOR MONITORING GOALS

Currently, Y-12 Plant surface water is not a source of public water before it reaches the Clinch River, where effluents are greatly diluted. Doses to the public have been determined through monitoring and assessment activities as reported in ASERs. No significant public doses have resulted from total releases from Y-12, the East Tennessee Technology Park, and ORNL.

Reference concentrations for Y-12 Plant radionuclides are given in Table 4. The Dose Conversion Factors (DCFs) are taken from EPA Federal Guidance Report 11. The 4 mrem/yr reference concentrations are obtained by dividing the DOE Derived Concentration Guides (DCGs) , which correspond to a 100-mrem/yr dose, by 25. The DOE DCGs are from DOE Order 5400.5. Also indicated in Table 4 are EPA proposed or established limits for specific radionuclides given in the ANPR (*Federal Register* 56, No. 138, July 18, 1991).

Use of secondary limits (e.g., concentrations) derived from general dose limits has been the standard practice of the ICRP, NCRP, the United Nations Scientific Committee on the Effects of Atomic Radiation, and other advisory bodies and regulatory agencies for at least 30 years. For more than one radionuclide, the usual practice is to sum site-specific doses and ensure that the total dose does not exceed the limiting dose. In terms of concentrations, the sum rule is applied such that if the sum of estimated concentrations (measured or calculated) divided by the limit for each radionuclide does not exceed unity, then the combined concentrations are within limits.
Thus, the sum rule for concentrations is:

$$\Sigma \mathrm{Ci/CLi} < 1,$$

where Ci is the concentration of radioisotope i in a particular environmental pathway and CLi is the concentration limit for radioisotope i in that pathway.

Use of the rule, taking into account dilution in the Clinch River for radioisotopes in Y-12 Plant surface waters and approximately accounting for other sources indicates that concentrations of about 0.2 DCG in East Fork Poplar Creek and Bear Creek would result in a total dose from direct consumption of Clinch River water of about 4 mrem/yr. (See Section 3.2 and Appendix A for further discussion). Therefore, MDAs of about 0.2 DCG or less are adequate for regulatory purposes.

Table 4. Dose Conversion Factors and Limiting Concentrations for Y-12 Radiological Contaminants

Nuclide	Ingestion Dose Conversion Factor (rem/µci)	4 mrem/year Concentration (pCi/L)	100 mrem/year Concentration (pCi/L***
U-234	0.28	19.0	500
U-235	0.27	21.0	600
U-238	0.25	22.0*	600
Th-228	0.40	14.0	400
Th-230	0.55	10.0	300
Th-232	2.70	2.0	50
Ra-226	1.30	4.1*	100
Ra-228	1.40	3.8*	100
Pu-238	3.20	1.7	40
Pu-239	3.50	1.5	40
Np-237	4.40	1.2	30
Am-241	3.50	1.5	30
Tc-99	1.46E-03	3.7E+03	1.00E+05
Sr-90	0.14	38.0**	1.00E+03
H-3	6.40E-05	8.6E+04	2.00E+06
Cs-137	5.00E-02	1.09E+02	2.60E+03

* EPA - proposed MCL is 30 pCi/L for uranium and 20 pCi/L for Ra-226 and Ra-228 (*Federal Register*, Vol. 56. No. 138 Thursday, July 18, 1991).

** EPA Safe Drinking Water Act (40 CFR 141.16) is 8 pCi/L for strontium, is 2E+04 pCi/L for tritium, and 9E+02 pCi/L for technetium-99.

*** From DOE Order 5400.5.

Typical MDAs reported by the Energy Systems Analytical Services Organization (ASO) are below the 4-mrem/yr concentrations (0.04 DCG) for each isotope (see Appendix A, Table 5). Therefore, they are more than adequate for regulatory purposes. An exception may be ^{90}Sr, where the regulatory level (8 pCi/L) is lower than the 4 mrem/yr concentration (38 pCi/L). Although the 8-pCi/L drinking water limit is much lower than the 0.2 DCG level (~200 pCi/L), the ASO has lowered the MDA for ^{90}Sr to 8 pCi/L. Such low MDAs are far below that necessary for regulatory purposes. Such low MDAs also provide a basis for confidently excluding radioisotopes with measured concentrations below monitoring goals from further NPDES monitoring.

The detection limits or MDAs necessary to meet monitoring goals based on DOE Order 5400.5 and draft Part 834 are given in Table 5.

The column entitled Current NPDES in Table 5 is based on the current DOE order and permit requirement that isotopes be monitored against DCGs. Since 16 isotopes are currently monitored, the monitoring goal for each isotope is 1/16 of its DCG from Table 4—a little higher than the 4-mrem/yr drinking water criterion (1/25 of the DCGs). These goals are adequate for regulatory purposes but not adequate to monitor current levels because current levels are often lower than these MDAs. (See Section 3.2). They are about three times lower than the 0.2 DCG level, based on a rough pathways analysis, discussed above and in Section 3 of Appendix A.

The practical quantitation level column lists the current MDA values which are reported by the Analytical Services Organization. They take into consideration current analytical laboratory capabilities without significant escalation in effort and cost.

The use of MDAs based on practical quantitation levels makes the data useful for a variety of purposes and is consistent with DOE and state wishes that data from on-going routine monitoring programs supplement environmental restoration investigations. They are well below current regulatory levels and take into account current analytical capabilities. Thus, they are probably ALARA although no formal ALARA analysis has been conducted. They are not necessary to demonstrate regulatory compliance but would reduce the need for special studies requiring MDAs lower than the regulatory requirements.

In summary, the Y-12 Plant must meet the "current NPDES" MDAs in Table 5 but can (and typically does) meet the practical quantitation levels.

Table 5. Surface Water Monitoring Goals for Y-12 Plant radionuclides

Monitoring Goals (pCi/L)

Parameters	Current NPDES[1]	Practical Quantitation Levels[2]
U-238	37.5	0.5
U-235	37.5	0.5
U-234	31	0.5
Th-228	25	0.5
Th-230	19	0.5
Th-232	3	0.5
Ra-226	6	1.0
Ra-228	6	3.0
Pu-238	2.5	0.5
Pu-239	2.5	0.5
Np-237	1.9	0.5
Am-241	1.9	0.5
Tc-99	6.25E+03	8.0
Sr-90	62.5	8.0
H-3	1.25E+05	400
Cs-137	1.60E+02	6.0

[1] Sum of fractions = 1 where limit is DCG for each isotope

[2] Typical detection limits currently reported by the Analytical Services Organization (See Table 5, Appendix A).

3.2 CONCENTRATION RATIOS FOR Y-12 RADIOISOTOPES

Station 17 (near the intersection of Bear Creek and Scarboro Roads) and Outfall 304 (near the intersection of Bear Creek Road and Route 95) represent the Y-12 Plant off-site surface water releases. Surface water concentrations of isotopes released from the Y-12 Plant at these locations are compared with their DCGs. At both locations, all isotopes except for ^{234}U and ^{238}U were at concentrations less than 1 percent of their DCGs. The isotopes ^{234}U and ^{238}U were at concentrations of about 1 percent of their DCGs. The sum of the percentages for all isotopes was about 3.3 percent at station 17 and about 4.8 percent at Outfall 304. Concentrations of 4 percent of DCGs are roughly equivalent to the 4 mrem/yr drinking water limits. Based on average annual flow rates for Bear Creek, East Fork Poplar Creek, and Clinch River, concentrations will be diluted by about a factor of 1000 upon mixing with the Clinch River. Thus, Y-12 Plant surface water releases will make a maximum contribution of 1/1000 of drinking water criteria via the drinking water route. Under current conditions, if concentrations are less than about 1,000/n times the drinking water criteria at the measurement point, then the total dose at the exposure point will not exceed 4 mrem/yr. (See Appendix A, Section 3).Taking into consideration releases from other sites and other pathways, releases from the Y-12 Plant should be monitored at levels of about 0.2 DCGs (See Section 3, Appendix A). The current regulatory column in Table 5 lists values of 1/16 of the DCGs, about a factor of 3 lower than the 0.2 DCG levels.

The highest levels of radioisotopes were at Y-12 Plant discharge point 512 (groundwater treatment facility) where the sum of ratios for uranium isotopes was about 6.2 percent and about 8 percent for all isotopes combined. The concentration of each isotope was compared with its DCG.

4. MONITORING METHODOLOGIES TO MEET EPA AND TENNESSEE REQUIREMENTS

In this section, the laboratory methods to be used to satisfy EPA and state of Tennessee requirements are given. These methods are supplemented when necessary to fulfill the more general requirements for consequence analysis (e.g., under the Resource Conservation and Reclamation Act [RCRA] or CERCLA) or research, based upon the January 1991 DOE *Regulatory Guide for Radiological Effluent Monitoring and Environmental Surveillance*. A summary of counting statistics as related to detection limits is also given.

The Y-12 Plant NPDES Permit (TN002968) contains the specific requirements of the state of Tennessee for surface water monitoring at Y-12. Monitoring goals under RCRA and CERCLA are being developed by the Environmental Restoration Program.

4.1 MONITORING METHODOLOGIES

The monitoring methodologies that will be utilized to monitor Y-12 Plant surface water and that will meet the monitoring goals discussed in Section 3 are given in Table 6. The NPDES Permit (Part III.H) requires the EPA methodologies. For the radiological monitoring component of the surface water monitoring program, the Y-12 Plant uses these methodologies and specific procedures and/or their equivalent or better as approved by the State.

Procedures for applying the methods listed in Table 6 may be found in the references following Table 6. The monitoring system can monitor to levels below the requirements of DOE Order 5400.5, evaluate trends, demonstrate that concentrations are ALARA, and provide a basis for an assessment of suitability for public use. Typical current MDAs, counting times, and sample volumes are given in Table 5 of Appendix A.

Table 6. Methodologies for Radionuclide Determinations

Radionuclide	Methodology	ASO Procedure	Reference
Gross alpha and beta	Evaporation/Gas Flow Proportional Counting	AC-LLL-EPA-600/900.0	EPA 900.0[1]
Total Alpha Radium	Barium Sulfate Coprecipitation Gas Flow Proportion Counting	AC-LLL-EPA-600/903.0	EPA 903.0[1]
Radium-228	Gamma Spectrometry	AC-LLL-EPA-600/901.1	EPA 901.1[1]
Americium-241	Neodymium fluoride coprecipitation and separation by chromatographic resin/ Alpha Spectrometry	AC-M-Z-22012 Y/P65-7157	2, 3, 4
Neptunium-237	Neodymium fluoride coprecipitation and separation by chromatographic resin/ Alpha Spectrometry	Y/P65-7206	3, 4, 5
Plutonium-238,239/240	Neodymium fluoride coprecipitation and separation by chromatographic resin/ Alpha Spectrometry	AC-MM-22012	3, 4, 6
Thorium-228,232,230	Neodymium fluoride coprecipitation and separation by chromatographic resin/ Alpha Spectrometry	Y/P65-7206	3, 4, 7
Uranium Isotopic	Neodymium fluoride coprecipitation and separation by chromatographic resin/ Alpha Spectrometry	AC-MM-22012	3, 4, 8
Total Uranium and % U-235	Solvent Extraction/Thermal Ionization Mass Spectrometry	Y/P65-7165	8
Cesium-137	Gamma Spectrometry	AC-LLL-EPA-600/901.1	EPA 901.1[1]
Strontium 89/90	Separation by carbonate coprecipitation ^{90}Y ingrowth, Gas Flow Proportional Counting	AC-LLL-EPA-600/905.0	EPA 905.0[1]
Technetium-99	Separation by chromatographic resin Liquid Scintillation Counting	Y/P65-7154	9
Tritium	Distillation/Liquid Scintillation Counting	AC-LLL-EPA-600/906.0	EPA 906.0[1]

Table 6 (cont.) References

1. Prescribed Procedures for Measurement of Radioactivity in Drinking Water, EPA Environmental Monitoring and Support Laboratory, Cincinnati, Ohio (EPA-600/4-80-032, August 1980).

2. Penneman, R.A., and Keenan, T. K.,*The Radiochemistry of Americium and Curium*, National Research Council, National Academy of Sciences NAS-NS-3006, January 1960.

3. Hindman, Forest D., "Actinide Separations for Alpha Spectrometry Using Neodymium Fluoride Coprecipitation, "*Anal. Chem*,. 58, 1986.

4. Hindman, Forest D., "Neodymium Fluoride Mounting for Alpha Spectrometric Determination of Uranium, Plutonium, and Americium," *Anal. Chem.*, **55**, 1983.

5. Burney, G.A., and Harbour, R.M., *The Radiochemistry of Neptunium*, National Academy of Sciences, National Research Council, NAS-NS-3060 December 1974.

6. Coleman, George H., *The Radiochemistry of Plutonium*, National Academy of Sciences, National Research Council, NAS-NS-3058, September 1965.

7. Hyde, E.K., *The Radiochemistry of Thorium*, National Academy of Sciences, National Research Council, NAS-NS-3004, January 1960.

8. Grindler, J.E., *The Radiochemistry of Uranium*, National Academy of Sciences, National Research Council, NAS-NS-3050, March 1962.

9. Anders, Edward., *The Radiobiochemistry of Technetium*, National Academy of Sciences, National Research Council, NAS-NS-3021, November 1960.

Specific isotopes currently determined by alpha spectrometry, if necessary, in Y-12 Plant surface waters include ^{241}Am,^{244}Cm,^{237}Np,^{238}Pu,^{239}Pu,^{240}Pu, ^{228}Th,^{230}Th,^{232}Th,^{234} U,^{235}U,^{236}U, and ^{238}U. Most of these do not need to be monitored under NPDES because their concentrations are well below "current NPDES" MDAs and even below practical quantitation MDAs (See Section 3.1). Typical sample size is 0.1 L. Counting time is 16.7 hours to achieve MDAs of less than 1 pCi/L. Specific isotopes currently determined by gamma spectrometry, if necessary, include ^{234}Pa, ^{137}Cs,^{60}Co,^{106}Ru. Typical sample size is 0.9 L. Counting time is 16.7 hours to achieve MDAs well below concentrations corresponding to a 4-mrem/yr dose. Concentrations of ^{137}Cs have been below a MDA of about 10 pCi/L compared with a 4-mrem/yr concentration of 110 pCi/L. ^{106}Ru concentrations have been below a MDA of about 40 pCi/L compared with a 4-mrem/yr concentration of 200 pCi/L. Gamma emitters have been below MDAs at the Y-12 Plant and thus do not make a significant contribution to TEDE under DOE Order 5400.5.

The only individual beta emitters currently monitored at the Y-12 Plant are ^{90}Sr,^{3}H, and ^{99}Tc.

The isotopes ^{3}H and ^{99}Tc are determined by liquid scintillation counting. Sample volumes are 0.05L for ^{3}H and 0.4L for ^{99}Tc. A counting time of 1 hour results in DLs of 400 pCi/L for ^{3}H and 8 pCi/L for ^{99}Tc, which are well below concentrations corresponding to a 4-mrem/yr dose (see Table 4). Although concentrations of these isotopes have been well below guidelines, surface water monitoring continues because they are regulated in drinking water supplies under the Safe Drinking Water Act and are detected in groundwater.

Similarly, ^{90}Sr is monitored because it is regulated under the Safe Drinking Water Act in drinking water supplies and occurs in groundwater. The strontium is precipitated and then counted using a gas flow proportional counter. A sample volume of 0.1 L and a counting time of 1 hour gives a detection limit of 15 pCi/L. This detection limit is below the 4-mrem/yr drinking water concentration of 38 pCi/L but is not adequate compared with the EPA MCL of 8 pCi/L (see Table 4). Therefore, the MDA for ^{90}Sr has been lowered to 8 pCi/L to meet the current EPA regulatory drinking water requirement.

4.2 DETECTION LIMITS AND COUNTING STATISTICS

The major implication of the monitoring requirements is that the MDA for each radioisotope must be below the monitoring goals appropriate to the application. The MDA as specified in the Safe Drinking Water Act is that concentration which can be counted with a precision of plus or minus 100 percent at the 95 percent confidence level. The basis for detection limits for the radioisotopes of concern in Y-12 Plant surface water are discussed in Section 3 and Appendix A.

Current methods of reporting monitoring results based on alpha spectrometry for Y-12 Plant surface water reflect subtraction of an "instrument background," R_B, value from the gross sample count, R(total), to yield a "net sample activity," R_S. Time is generally held constant and the "percent error" is allowed to vary. Subtraction of R_B from R(total) can lead to negative numbers with large errors, especially if R_B does not include the same matrix effects as R(total).

Standard counting statistics involve determination of sample and instrument background counting times to achieve a desired counting error. The standard procedure is in accord with EPA and DOE requirements. Under ideal conditions, "counting error" is determined in the laboratory using known sample activities and counter backgrounds. Under practical conditions MDA will necessarily be different than under ideal laboratory conditions. The total counting time T is given by:

$$T = [\ R(\text{total})^{1/2} + {R_B}^{1/2}\ ]^2 \,/\, V^2 {R_S}^2,$$

where

$$V_i = 100\ \sigma_\iota / P\iota$$

is the coefficient of variation (percent error) of R_i (count) and s_i is the standard deviation of R_i. Clearly, counting times can become very long for low R_S values at a given percent error. MDAs are a function of counting times and desired percent error. Larger sample volumes yield higher sample count rates so that desired percent errors can be achieved with shorter counting times. Ultimate sensitivity resolves to practical technical and economic issues. In the equations, R_B is instrument background, usually well defined, whereas R(total) and R_S in environmental water samples incorporate not only instrument background but also environmental background, R_E, which for natural radionuclides (e.g., uranium) is highly variable.

$$R_{SB} = R_S + R_E + R_B.$$

The net sample count is therefore not simply R(total)-R_B but R(total)-R_E-R_B. To determine R_E, background samples must be supplied to the analytical laboratory. If environmental background is based on an average value, as it practically must be, then normal variation in environmental background (at least an order of magnitude) will produce variation in net results, which, if near environmental background, will produce about as many negative as positive results when the average environmental background is subtracted. A negative net count simply means that the sample result is within the combined normal variation in background and variation in R_S. If a negative count results from subtraction of counter background only from total count, then matrix effects, counter contamination, interference—along with inadequate detection limits, are probable causes.

Radiological data in the Environmental Monitoring Management Information System (EMMIS) will include counting error determined by standard techniques incorporated into the software associated with the analytical instruments used by the ASO. Methods used by Energy Systems for determination of MDAs are those recommended in ANSI N13.30, *American National Standard for Performance Criteria for Radiobioassay*, and by the EPA. Concentrations of alpha-, gamma-, and beta-emitting radionuclides in Y-12 Plant surface waters are generally low compared with a variable environmental background. Use of average values for backgrounds may lead to special radiological, statistical, and analytical problems that require expert analysis. These analyses will be conducted as necessary.

Examples of such analyses requiring radiological or health physics expertise are given in Section 5.2 and Appendix A.

5. DATA MANAGEMENT AND RADIOLOGICAL ANALYSIS

5.1 DATA MANAGEMENT

All environmental data gathered by the Compliance Monitoring Section of the Y-12 Plant Environmental Management Department will be entered directly by the analytical laboratory into the Energy Systems Laboratory Information Management System (ESLIMS) on the same day it is obtained. ESLIMS will be electronically accessed on a routine basis by EMMIS. EMMIS will capture pertinent data in ESLIMS as a result of sample initiation occurring in EMMIS. This system will store the data generally in the form of concentrations in various media. EMMIS will compare the concentrations with regulatory and best management or ALARA goals and produce required reports. The state of Tennessee requires that initial comparisons be made with DCGs. Radiological data, in addition to other types of data, are also transferred into EMMIS where they are readily retrievable for use in reports or special statistical analysis.

5.2 RADIOLOGICAL ANALYSIS METHODOLOGY WITH EXAMPLES

The Y-12 Plant surface water radiological databases have been analyzed (1) to support update of the Radiological Monitoring Plan and (2) as part of a broader effort to assess whether the current radiological water monitoring program is effectively monitoring radiological contamination produced by Y-12. A general analysis is given Appendix A. A summary with examples based on recent monitoring results is given here.

Activities at the Y-12 Plant primarily involved uranium processing at various levels of enrichment in ^{235}U. Radioisotopes to be considered from these activities are ^{238}U, ^{235}U, and ^{234}U. In addition, some recycle uranium from reactor fuel elements containing fission products and transuranics have been processed. Also, some ORNL activities at the Y-12 Plant involved fission product radioisotopes. ORNL also conducted biological research that utilized fission products, transuranics, and accelerator-produced tracers. Some thorium processing was conducted at the Y-12 Plant, but thorium from reactor fuel elements was not processed.

On the basis of general Y-12 Plant operational history, water monitoring at the Y-12 Plant has included alpha, beta, and gamma screening; the uranium isotopes ^{234}U, ^{235}U, and ^{238}U; weight percent enrichment of ^{235}U; the fission and activation products ^{60}Co, ^{137}Cs, ^{3}H, ^{90}Sr, and ^{99}Tc; the transuranics ^{241}Am, ^{237}Np, ^{238}Pu, and ^{239}Pu; and biological tracers such as ^{14}C, ^{35}S, ^{32}P, ^{129}I, and ^{125}I. Isotopes monitored varied from site to site because the activities potentially releasing radioisotopes to those sites varied.

Operational history and monitoring results have provided a basis for reducing the general lists of radioisotopes monitored at the Y-12 Plant to ^{234}U, ^{235}U, ^{238}U, ^{237}Np, ^{241}Am, ^{228}Ra, ^{226}Ra, ^{228}Th, ^{230}Th, ^{232}Th, ^{234}Th, ^{99}Tc, ^{90}Sr, and ^{3}H in addition to alpha and beta screening measurements. Total uranium and weight percent ^{235}U are also monitored. The 1991–1994 results for selected outfalls had been re-evaluated previously. The 1995 results are used here to illustrate the general analysis methodology and improvements that have resulted. The basis for the current monitoring program is discussed in more detail in Appendix A.

In addition to continued comparison of the data with reference standards for each outfall, the data are analyzed to check whether results are consistent with physical and chemical expectations and plant operational history. For example, the relationships among radioisotopes in a decay chain are predictable if no unusual sources for a particular isotope exist in the chain, if the time history is known, and if no differential removal or concentration mechanisms for a particular isotope exist.

If the results are not in agreement with expectations from known relationships, plant operations, and pathways analysis, those results are investigated further. If no explanation can be found for the unexpected results, they are considered suspect until they are confirmed by repetition. Confirmed results not in agreement with expectations stimulate investigation of the entire sample collection and analytical laboratory system. Process variables may also be reviewed.

For the radioisotopes monitored at the Y-12 Plant, some of the relationships expected and their application were described in the 1995 version of the RMP. This analysis will focus on NPDES requirements with respect to gamma scans and beta emitters in surface water. A summary of selected comments and responses from recent audits on sanitary sewer and settleable solids monitoring will also be given.

5.3 REQUIREMENTS OF THE NPDES PERMIT

The Y-12 Plant NPDES Permit requires an assessment and re-evaluation of the RMP, which may provide a basis for refinement and/or modification of the RMP. The assessment of the 1995 version of the RMP included an evaluation of initial gamma scanning results and data collected according to the Storm Water Pollution Prevention Plan.

The NPDES Permit did not specify requirements for related sanitary sewer or settleable solids/sediment monitoring. However, brief discussions are given here because monitoring is required by DOE Orders and some questions arose about these monitoring activities during recent audits.

General Review of Data

The 1995 data on concentrations of radioisotopes at Y-12 Plant surface and sanitary sewer water is generally consistent with data from previous years on a monthly or annual average basis. As expected, uranium dominates radiological releases based on comparison of concentrations to DCGs. However, there were some unusual spikes in the weekly data for total uranium in 1995 compared with 1993 and 1994 data. Large differences between calculated total uranium based on mass spectrometry (MS) and based on alpha spectrometry (AS) are being reviewed to identify causes. Agreement between calculated total uranium based on MS and AS was generally good, except for the peaks. Since the MS and AS data are for the same samples, it is likely that the differences result from sampling or analytical differences rather than from a separation of isotopes in water.

Based on a review of the surface water data, higher flow rates at Stations 8 and 17 result in higher monthly average concentrations. Therefore, uranium is mobilized by storm water flow. Otherwise, with a relatively constant uranium source, uranium concentrations would show a decreasing trend with increasing flow rates. This mobilized uranium could result from general surface runoff or from the suspension of previously contaminated sediments during storm flow. Some contaminated ground water could also be flushed out during storm events. Some information on possible locations of sources can be obtained from the data.

The concentrations of uranium at Station 17 are predictable based on concentrations at Station 8 multiplied by the ratio of flow rate at Station 8 to the flow rate at Station 17. One interpretation of these results would be that no significant sources of uranium appear to be mobilized between Station 8 and Station 17 and that no significant deposition of uranium occurs between Station 8 and 17. Another, but less likely, interpretation would be that uranium is mobilized but redeposited between Stations 8 and 17. The initial storm water data confirms that only low levels of uranium enters EFPC between Stations 8 and 17.

In Bear Creek Valley, uranium is mobilized between BCK 11.97 and Outfall 304 by surface runoff. Runoff surface water concentrations in Bear Creek (BC) are insensitive to flow rates. At BCK 11.97, the concentrations were about the same (102 and 103 pCi/L) even though the flow rate increased by a factor of 1.76 from 1993 to 1994. At Outfall 304 the flow rate increased from 6.87 to 12 cfs (1.75x) from 1993 to 1994, but the concentration only decreased from 14.6 to 13 pCi/L. Based on flow rate ratios at BCK 11.97 and Outfall 304, the uranium concentration at Outfall 304 would be about 6 pCi/L. The increased uranium mobilized by storm water flow appears to be diluted by the increased flow so that uranium concentration at Outfall 304 is essentially unchanged.

As discussed above for EFPC, it is possible that increased flow from storm events mobilizes creek sediments with relatively high concentrations of uranium. However, sediment measurements in both Bear Creek and East Fork Poplar Creek yield concentrations no more than twice the normal soil background values (See Section 5.5). Therefore, the increased releases from storm events probably result from runoff from contaminated surfaces and groundwater flow. At base flow conditions, contaminated groundwater is the only significant source for contaminants in BC and EFPC. A review of the initial storm water data indicates that most of the uranium from storm water flow in BC originates from groundwater flow from the capped disposal area at the west end of the Y-12 Plant site.

An indicator of contamination from groundwater other than uranium is ^{99}Tc. The concentration of ^{99}Tc at Outfall 304 is much more closely predicted by the flow rate ratio for BCK 11.97 and Outfall 304. This indicates that ^{99}Tc had entered BC before BCK 11.97. The concentrations of ^{99}Tc at BCK 11.97 and Outfall 304 are relatively unaffected by higher flows. Therefore, enough additional ^{99}Tc enters Bear Creek from groundwater from storm events to offset the dilution potential of greater flows.

Increased uranium during storm flow probably results from surface sources as well as from increased groundwater flow. Additional storm water monitoring data should help to further define relative contributions.

Preliminary Review of Gamma- and Beta-Emitter Data

Given that more than 10 years has elapsed since recycle uranium was last processed at the Y-12 Plant, the only process-associated isotopes expected to be detected by gamma spectroscopy are ^{137}Cs and, perhaps, ^{145}Pm, ^{146}Pm, ^{150}Eu, ^{152}Eu, and ^{60}Co (See Table 2, Appendix A). Only ^{137}Cs and ^{60}Co were measured.

Gamma scanning results through June 1996 confirm the analysis given in Appendix A. At Outfall 200, which represents the highest concentrations of radioisotopes in EFPC, ^{137}Cs concentrations exceeded an MDA of about 3 pCi/L 5 times out of 13 measurements. The highest value was 8.5 pCi/L compared with a DCG of 3,000 pCi/L. The concentrations of ^{60}Co exceeded MDAs ranging from about 3 to 5 pCi/L 4 times out of 13 measurements, but two of these values were estimated. The highest ^{60}Co concentration was 10 pCi/L compared with a DCG of 5000 pCi/L. All other fission or activation product gamma emitters had reported concentrations less than their respective MDAs. None of the reported gamma-emitting isotopes other than ^{137}Cs and ^{60}Co were expected to have concentrations greater than about 1% of the ^{137}Cs concentration or greater than about 0.1 pCi/L (see Appendix A). Other gamma-emitting fission or activation products that are produced in quantities up to 10% of ^{90}Sr (Appendix A) and have sufficiently long half-lives were not measured (e.g., ^{145}Pm, ^{146}Pm, $^{150}E_{u,}$ $^{152}E_{u}$). However, their hazard potential relative to ^{90}Sr is very low.

Among the fission product beta emitters, the isotope that might exhibit the highest concentration, ^{90}Sr, had no values above the MDA at Station 200. The MDA ranged from about 8 to 11 pCi/L, roughly 1% of the 1000 pCi/L DCG. The fission products appear to originate from the Groundwater Treatment Facility (represented by Outfall 512), which may more efficiently remove ^{90}Sr compared with ^{137}Cs. Other beta-emitting fission or activation products may have concentrations in fuel elements comparable to ^{90}Sr (e.g., ^{93m}Nb), but their hazard potential relative to ^{90}Sr is very low.

The other isotopes monitored since July 1995 under the new permit are not expected to be present. ^{22}Na, ^{51}Cr, ^{54}Mn, ^{57}Co, ^{58}Co, ^{59}Fe, ^{65}Zn, ^{95}Nb, ^{95}Zr ^{106}Ru, ^{134}Cs and ^{144}Ce all have very short half-lives relative to 30 years. ^{125}Sb has a 2.7- year half-life, so 30 years represents about 10 half-lives. ^{208}Th, ^{212}Bi, and ^{212}Pb are in the $_{232}$Th series and would not be expected to be elevated unless $_{232}$Th is elevated. The large difference between ^{235}U based on alpha spectrometry and gamma spectrometry may result from the higher MDA for gamma spectrometry or interferences from other natural gamma-emitting radioisotopes. Appendix A provides a discussion of gamma emitters associated with processing of recycle uranium, including monitoring recommendations.

5.4 SANITARY SEWER SYSTEM

Uranium Isotopes

As in previous years, releases of radioisotopes via the sanitary sewer system at the Y-12 Plant are dominated by uranium isotopes. Differences between analyses based on AS versus MS persist. Most of the differences between AS and MS are accounted for by ^{234}U, where a relatively small mass change leads to a relatively large activity change. However, very large differences between MS and AS occurred (e.g., August 29, 1995, and January 9, 1996) even though ^{238}U and ^{234}U were about the same. Several other samples exhibited relatively high AS results, often with no equivalent increase in MS results. It should be noted that these relatively high results were only a fraction of the relevant DCGs.

Gross alpha, beta, and gamma measurements are not necessary since the dominant isotopes associated with the Y-12 Plant sewer system are measured. No correlation between gross alpha, beta or gamma and the uranium isotopes is evident. Beta and gamma emitters expected to be in equilibrium with ^{238}U (^{234}Th and ^{234m}Pa) are not reflected in the gross measurements, even for relatively high levels of the isotopes. Similarly, relatively high levels of ^{238}U and ^{234}U are not reflected in the gross alpha results. Gross alpha, beta, and gamma levels are, therefore, not reliable as screening procedures to indicate the need for specific isotope measurements. Conversely, gross alpha levels above some screening levels; e.g., 15 pCi/L, would have led to unproductive searches for the associated isotopes.

Reasonably good correlation between gross alpha measurements and total uranium has been observed but usually at much higher levels than typical of the Y-12 Plant releases. In most cases, the levels were much higher than highly variable background levels. For the usual low levels encountered at the Y-12 Plant, it is necessary to continue measurement of specific isotopes. MS measurements should also continue (total uranium and mass percent ^{235}U). Until the differences between MS and AS can be explained, both methods should be used.

Concentrations of total uranium in Y-12 Plant wastewater released to the Oak Ridge Sanitary Sewer System are less than drinking water criteria. The 1995 annual average concentration of total uranium in Y-12 Plant wastewater was 8.71 pCi/L based on AS and 3.53 pCi/L based on MS. The EPA MCL is 30 pCi/L for total uranium (*Federal Register*, Vol. 56, No. 138, Thursday, July 18, 1991). The limits in DOE Order 5400.5 are concentrations limits at sewer release points of five times the DCGs, annual total releases of 5 Ci for ^{3}H, 1 Ci for ^{14}C, and 1 Ci for others combined. Potential public doses must be a small fraction of the primary EDE limit of 100 mrem/yr. On an annual average basis the Y-12 Plant wastewater system clearly meets these limits by a wide margin.

The 1995 annual average level is higher than the 1994 annual average level. The higher 1995 level resulted from a short-term release associated with sewer line inspection (see below).

On-Site Sources of Uranium in the Y-12 Plant Sanitary Sewer System

Although the Y-12 Plant contribution of uranium to the Oak Ridge Sanitary Sewer System is very low, the DOE Office of Nuclear Safety (U.S. DOE, 1993) commented that DOE/Oak Ridge Operations (ORO) had not fully quantified the concentration of uranium entering the sanitary sewer system from known sources such as hand wash basins and dishwashers at the Y-12 Plant.

Additional sampling has been conducted at several locations within the Y-12 Plant sanitary sewer collection system. Sampling included the sanitary sewer lines from a change house and labware dishwasher. Releases to the sanitary sewer system from a hand washing station used to remove low level uranium contamination were estimated by Y-12 Plant health physicists to be less than 0.2 microcurie per year. Such releases would result in concentrations of less than 2×10^{-4} pCi/L at the East End Monitoring Station. Sampling also included sewer mains that serve several different areas of the Y-12 Plant.

Total uranium mass values and isotope uranium values for sanitary sewer lines at the west end of the Y-12 Plant ranged from less than 2 to 54 ppb and from 5 to 30 pCi/L with percent mass of ^{235}U varying from about 0.4 to 5%. While these concentrations are less than drinking water criteria (see Table 2), they are higher than expected background concentrations. Concentrations in the wastewater from the labware dishwasher were in the low end of the range. Total uranium concentrations in wastewater from the change houses were consistently less than 1 ppb. Specific sources are not identifiable from the data.

Since portions of the sewer line are old and probably leak, they could receive surface water and groundwater. Concentrations of uranium isotopes in the surface water in the vicinity of the sewer line samples are in the same range as the sewer line samples. The highest groundwater concentrations at the west end of the Y-12 Plant are associated with S-3 plume. Although the S-3 site is west of the hydrologic divide that separates the Bear Creek Regime from the East Fork Poplar Creek Regime, the S-3 plume has crossed the divide because of a mound in the water table created by the disposal of large volumes of liquid wastes during operation of the S-3 site. The sewer samples were taken in the western end of the East Fork Poplar Creek regime. There the S-3 plume mixes with other plumes from sites in the East Fork Poplar Creek regime.

A study of the Y-12 Plant Sanitary Sewer System was conducted in 1995 in preparation for the sanitary sewer upgrade project to repair defective sewer lines. This study involved inspection of the pipelines using a video camera. Before video inspection, high-pressure water cleaning of the pipes is necessary. During this inspection, sediment material in the sewer system was mobilized and released throughout the sanitary sewer system. One weekly sample (August 29, 1995) had a total uranium level at the EEMS of 157 pCi/L. Although this level is well below the 5 times DCG limit given above, it is about 20 times above normal levels. As sewer lines are repaired or replaced, external sources of contamination will be eliminated. To prevent additional releases of contaminants, all wastewaters associated with the sanitary sewer upgrade project will be collected until monitoring confirms the concentrations of contaminants are acceptable for discharge.

Plutonium Isotopes

It is doubtful that continued monitoring for ^{238}Pu and $^{239/240}$Pu is useful. Reported levels have been below MDAs, which are below drinking water criteria. The uncertainty associated with the data is very high. The plutonium isotopes also do not exhibit increases even when the sanitary sewer system is disturbed and uranium increases over the short term. These results imply that the plutonium isotopes are not present in and around the sewer system at levels that can be measured with desired accuracy using adequately low MDAs.

Based on data for a number of years for the sanitary sewer system, surface water and groundwater, general monitoring for plutonium isotopes in the sewer system should be discontinued. Some monitoring could be focused on suspected sources, if any.

5.5 LOCKHEED MARTIN CORPORATE AUDIT COMMENTS ON DOE REQUIREMENTS FOR SETTLEABLE SOLIDS IN LIQUID RELEASES

The DOE requirement that stimulated questions during the corporate audit is contained in DOE Order 5400.5, Chapter II, 3.a. (4), "Sedimentation." The requirement is:

"To prevent buildup of radionuclide concentrations in sediments, liquid process waste streams containing radioactive material in the form of settleable solids may be released to natural waterways if the concentration of radioactive material in the solids present in the waste stream does not exceed 5 pCi (0.2 Bq) per gram above background levels of settleable solids for alpha-emitting radionuclides or 50 pCi (2 Bq) per gram above background levels of settleable solids for beta-gamma-emitting radionuclides."

Settleable solids means those solids suspended in waste water that are determined to be settleable using Method 209E, Settleable Solids, pp. 98 and 99, *Standard Methods for Examination of Water and Waste Water,* 16th edition. This method has a practicable volume detection limit of about 0.1 milliliters per liter of water.

DOE Order 5400.5 does not discuss exceptions or alternatives when settleable solids are below reasonable volume detection limits of Method 209E.

To help determine whether concentrations of radioisotopes in settleable solids (SS) in releases to surface water from the Y-12 Plant might exceed detection limits, samples were collected from the Y-12 Plant waste streams. Values for SS in waste streams were less than the 0.1 mL/L detection limit.

Concentrations of total suspended solids (TSS) in releases to surface waters at the Y-12 Plant are also generally less than the TSS detection limit (5 mg/L). TSS may represent an upper limit for SS. Given that uranium isotopes are the dominant alpha-emitting radioisotopes at the Y-12 Plant, they can be used to examine whether alpha-emitting radioisotopes in SS can be monitored using practicable detection limits.

Using TSS at 5 mg/L as a limit for SS, the concentration of uranium in SS at the DOE 5400.5 limit of 5 pCi/g would be 0.025 pCi/L. All the TSS in a 40-L water sample would provide 1 pCi of uranium alpha activity, which is a practicable amount for reasonable counting times. Since the SS component is probably less than TSS, actual sample volumes would have to be substantially greater than 40 L to obtain 1 pCi of activity in SS.

By contrast, a few grams of sediment would provide several pCi of uranium alpha activity at background concentrations.

Therefore, at the Y-12 Plant, general periodic sediment sampling is preferred for practically attainable results to ensure that current operations are not contributing to the buildup of residual radioactive material in sediments.

Given the practical limitations on determining radioisotope concentrations in SS in Y-12 Plant streams, determination of the sources for any excess sediment concentrations may be difficult. Several general "open issues" arise in connection with interpretation of sediment results:

- What is an appropriate limit on sediment concentrations under the existing DOE Order 5400.5?
- What is the "significance" of the Y-12 Plant contribution to sediment concentrations?
- What is the relationship between sediment and SS concentrations?
- What is the relationship between the DOE order radioisotope limits and the volumetric detection limit (mL/L) of the analytical method used for SS?

Considerable effort would be required to substantially resolve these "open issues." They can be discussed only in a general way here to clarify whether they are vital issues that must be resolved before any acceptable analysis with respect to sediment concentration can be offered.

Limits

Since most of the sediment in BC and EFPC results from general surface runoff, appropriate background values for sediments might be normal soil background values. Since the intent of DOE Order 5400.5 limits for SS were to prevent buildup of radioisotopes in sediments, sediment concentrations above background would require investigation.

Although sediment background values for the Y-12 Plant are not available, background soil values for the Oak Ridge area are available from related studies. A study to obtain background values for heavy metals, including uranium, was conducted by Oak Ridge Associalted Universities (ORAU). A summary of the results was provided by Clayton Gist of DOE/ORO. Based on 46 samples of the Oak Ridge area background, uranium has a mean value of about 3.5 µg/g. The minimum value was 2.3 µg/g, the maximum value was 5.4 µg/g, and the standard deviation was 0.72 µg/g. If values more than two standard deviations above the mean value are taken to be "above background," then total uranium above about 5 µg/g in soil is considered "above background."

For soils that have not been disturbed, the specific activity of total uranium is about 0.67 pCi/µg. Therefore the total uranium activity concentration that might be considered above background is 5 µg/g × 0.67 pCi/µg ~ 3.3 pCi/g. For natural uranium, ^{234}U, ^{230}Th, and ^{226}Ra are expected to be approximately in secular equilibrium with ^{238}U. In contrast, the refined uranium processed at the Y-12 Plant is expected to be low in ^{230}Th and ^{226}Ra because they would have been separated from uranium in other components of the fuel cycle. Isotopic ratios possibly serve as an indicator of Y-12 Plant contributions to sediment concentrations. If the ratios are typical of natural uranium, the Y-12 Plant waste streams are not a strong contributor. Results will often reflect a mixture of natural uranium and refined uranium.

In a more recent study, *Final Report on the Background Soil characterization Project at the Oak Ridge Reservation, Oak Ridge, Tennessee*, the overall median concentration for total uranium was 2.44 pCi/g and the upper 95% confidence level value for the median was 2.84 pCi/g. The overall average estimate for the 95th percentile was 4.336 pCi/g, but this estimate included values for ^{235}U that appear to be high. A better estimate is about 4 pCi/g. The overall lower tolerance based on the 95th percentile estimate was 3.6 pCi/g. Values above 3.6 pCi/g are taken to be above background with 95% confidence. Again, the ^{235}U values were high. The corrected value would be about 3.4 pCi/g. The uranium isotopes exhibited near equilibrium values while ^{230}Th and ^{226}Ra exhibited values between 60 to 100% of equilibrium. Although the values used here might be adjusted through more detailed study, they are adequate for this preliminary qualitative analysis. Values above 3.4 pCi/g may be considered to be above ORR background values under DOE Order 5400.5.

Significance of Y-12 Plant Contributions to Sediment Concentrations

The total dose through all pathways from background equilibrium concentrations of ^{238}U decay chain isotopes in soil averages about 220 mrem/yr for indoor exposures (NCRP, 1987). If the variation in background doses reflects the variation in background soil concentrations, then the standard deviation in the dose would be about 45 mrem/yr. Uranium isotopes alone (^{238}U and ^{234}U) contribute only about 0.6 mrem/yr to the average. Most of the dose (~200 mrem/yr average) comes from indoor concentrations of radon and short-lived daughters that arise from ^{226}Ra concentrations in soil.

The doses from radon daughters, however, vary more widely than implied by soil ^{226}Ra values because ^{222}Rn is affected by many geological, meteorological, and building variables. A lognormal distribution with a geometric standard deviation of about 3 is probably appropriate.

Since the Y-12 Plant processes refined uranium containing little ^{230}Th or ^{226}Ra, or other daughters, its primary contribution is uranium isotopes only. Since ^{230}Th has a half life of about 75,000 years, it would take several hundred thousand years for ^{230}Th (and daughters) to reach equilibrium with the uranium isotopes. Sediments are likely to be removed and replaced many times during such time periods. Therefore, doubling uranium content of sediments due to releases from the Y-12 Plant would increase average background doses by a maximum of about 0.6 mrem/yr even if scenarios exist that would complete all possible exposure pathways (food, water, external, inhalation).

An increase in uranium concentration to twice background activity would not represent a significant increase in sediment concentrations If all pathways are not complete or, as expected at the Y-12 Plant, only a few if any, are complete, uranium concentrations could be much higher without significantly increasing human doses.

Relationship Between DOE Radioisotope Limits and SS Detection Limits

If sediment radioisotope concentration limits (when established) are exceeded and if isotopic ratios indicate a possible Y-12 Plant source, then it would be necessary to identify specific sources. The SS detection method specifying concentration in mL/L is not adequate because at the 0.1 mL/L volume detection limit, SS radioisotope concentrations could exceed the DOE limits for settleable solids in pCi/g. In order to confirm compliance with the DOE radioisotope limits, radioisotope concentrations in SS would have to be determined. However, the DOE limits apply for waste streams that are not likely to be high in SS. More likely sources might be surface, waste site or general contamination mobilized by construction or remediation activities or storm water. The storm water monitoring plan should provide additional information concerning whether and how much storm water releases might be contributing to sediments. Other contributors should be only occasional, temporary contributors.

Summary of Sediment Measurements

Results for 1993–1995 are listed in Table 7. Uncertainties in the numbers range from 5% for the higher values (^{238}U and ^{234}U) to 20% for the lower values (^{235}U). Based on comparison of ^{234}U and ^{238}U values, which are about the same for natural uranium, the uranium in sediments may be approximately classified as follows:

EFPC 23.4 - natural uranium ratios
EFPC 5.4 - slightly enriched
BCK 9.4 - slightly depleted
BCK 0.6 - slightly depleted

Any Y-12 Plant waste stream contributions to the sediments are expected to be depleted in ^{230}Th and ^{226}Ra because they were removed in other components of the fuel cycle for Y-12 Plant process uranium. The ratios of ^{230}Th to ^{238}U in the sediment samples are approximately:

EFPC	23.4	100%	(1993)	BCK	9.4	1%	(1993)
		24%	(1994)			4.2%	(1994)
		57%	(1995)			17.5%	(1995)
						6.3%	(1995)

EFPC 5.4	45%	(1993)	BCK 0.6	3.5%	(1993)
	22%	(1994)		12.8%	(1994)
	26%	(1994)		23 %	(1995)
	12.5%	(1995)			

Although these ^{230}Th to ^{238}U ratios are highly variable, they indicate that a larger fraction of the uranium in EFPC is from natural uranium than is the case in BC. Uranium in BC comes primarily from groundwater.

Data for ^{226}Ra were not reported.

Table 7. Sediment Concentrations of Radioisotopes Near the Y-12 Plant

		Isotope Concentration (Bq/kg)[1]			
Location	Date	U-234	U-235	U-238	Total U[2]
EFPC 23.4	8/06/93	100	4.1	88	192.1
	9/19/94	110	5.4	110	215.4
	10/09/95	110	4.3	120	234.3
EFPC 5.4	10/13/93	43	3.5	40	83.5
	9/19/94	87	5.7	60	152.7
	9/19/94	58	2.5	53	113.5
	10/10/95	130	6.0	88	224.0
BCK 9.4	10/15/93	69	5.7	93	167.7
	9/19/94	78	3.2	120	201.2
	10/09/95	45	2.1	80	127.1
	10/09/95	62	3.1	110	175.1
BCK 0.6	10/15/93	47	4.5	78	130.5
	9/19/94	14	0.62	21	35.6
	10/09/95	20	1.7	34	55.7

[1] Multiply Bq/kg by 0.027 to obtain pCi/g.

[2] Total uranium in background soils is about 85 Bq/kg average. Average plus 2 standard deviations is about 120 Bq/kg, based on Oak Ridge area soils (see text).

Based on the data for 1993, 1994, and 1995 (Table 7), the highest sediment uranium values (EFPC 23.4) are about 2× background, or about 2.5 - 3 pCi/g excess over background for soil.

However, based on the isotopic ratios, the uranium probably comes more from natural sources rather than from the Y-12 Plant waste streams.

The values at EFPC 5.4 were "above background" for one value in 1994 and for 1995. The uranium is slightly enriched. The sediments have a lower $^{230}Th/^{238}U$ ratio than for EFPC 23.4. The sources for the uranium at EFPC 5.4 probably do not include current Y-12 Plant waste streams because of distance, different enrichments and isotopic ratios, and other possible sources along EFPC. During earlier operational periods, significant quantities of uranium were released to EFPC from different processes. Part of this uranium was deposited along EFPC and could be periodically mobilized in EFPC waters to settle out further downstream.

The uranium values at BCK 9.4 were "above background." The uranium was slightly depleted. The ratios of ^{230}Th to ^{238}U were low. Thus, excess uranium in sediments at BCK 9.4 is probably from the Y-12 Plant. However, since the Y-12 Plant does not currently discharge any waste streams directly to BC, it is probably due to groundwater releases from waste sites rather than from current waste streams. Storm water data (Section 5.3) indicate releases from the capped West End disposal areas.

Uranium concentrations at BCK 0.6 were "above background" for 1993 but significantly below background for 1994 and 1995. The uranium was slightly depleted. The $^{230}Th/^{238}U$ ratios were low. According to these characteristics, some of the uranium at BCK 0.6 could come from the Y-12 Plant. It is not clear why the values for 1994 and 1995 are below soil background values.

None of the sediment uranium concentrations represent "significant" increases above soil background values. Therefore, the "open issues" discussed above currently are not vital with respect to interpreting sediment results. Although a pathways analysis has not been done, it is certain that not all of the pathways associated with background concentrations of natural radionuclides are complete for the Y-12 Plant sediments. Actually, if the sediments depleted in ^{230}Th were mixed with normal soils with equilibrium concentration of ^{238}U daughters, the mixture would also be depleted in ^{230}Th. Until the ^{230}Th reaches equilibrium (several hundred thousand years), there would be a net reduction in dose, compared with background dose, from ^{230}Th. Depletion in ^{226}Ra would make a much larger difference due to the major contribution of its daughter, ^{222}Rn, to background doses. If the ^{226}Ra concentrations in amended soils were reduced by a factor of 2 (assuming such a scenario occurred), the projected initial dose from radon and short-lived daughters would be reduced by 100 mrem/yr for indoor exposures. A detailed pathways analysis, if conducted, should include isotopes that are depleted as well as those that are enhanced. ^{226}Ra should be added to the list of isotopes measured in sediments.

Conclusions

1. Sediment measurements at the Y-12 Plant should continue because SS concentrations for the method used are below the detection limit. The relationships among SS concentrations in mL/L and radioisotope concentrations levels in DOE Order 5400.5 are not defined. The relationship may be difficult to define because SS samples are below the 0.1 mL/L detection limit.

2. Sediment concentrations are currently low and do not represent a "significant" increase above background. Continued sediment monitoring to recognize trends should satisfy the intent of DOE Order 5400.5.

3. Currently, pathways analyses are not considered necessary. If pathways analyses are conducted to consider some hypothetical future use scenarios, they should include isotopes that are depleted as well as those that are enhanced compared to natural uranium ratios in Y-12 waste streams.

6. QUALITY ASSURANCE

In May 1995, DOE issued as a controlled document the Environmental Monitoring Plan for the Oak Ridge Reservation (DOE/OR, 1995). Chapter 9 of this document is entitled Quality Program Plan. This chapter was developed in compliance with DOE Order 5700.6C, "Quality Assurance", American Society of Mechanical Engineers (ASME) "Quality Assurance Program Requirements for Nuclear Faculties" (NQA-1), and guidance listed in the DOE Order 5400.1 and the Regulatory Guide. In addition, this program was developed with Energy Systems QA requirements compatible with QA document Y/QD-15 Rev. 1. The TDEC DOE Oversite Organization contributed review and comments to an early draft of the document. These comments were considered and incorporated as appropriate.

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Appendix A

OVERVIEW RADIOLOGICAL ANALYSIS OF THE Y-12 PLANT SURFACE WATER RADIOLOGICAL MONITORING PROGRAM

1. INTRODUCTION

In a May 4, 1995, memo, the state of Tennessee Division of Water Pollution Control stated their needs and goals for radiological monitoring plans (RMPs), associated documentation, and data resulting from the RMPs required under the NPDES permitting system.

The memo stressed the following:

- Monitoring should be conducted at those outfalls that have the potential to discharge radioactivity to waters of the state.

- Information about radiological parameters and results should be made available to the public.

- RMPs should be part of an overall site effort to quantify radiological releases, radionuclide transport, determine background concentrations, and identify trends.

- Radiological monitoring under NPDES should be consistent with DOE requirements in DOE Orders and upcoming regulations under 10 CFR Part 834.

- Storm water monitoring should be incorporated into NPDES RMPs.

- RMPs or associated documentation should provide the basis for the radiological parameters in the RMPs.

- RMPs should specify the monitoring goals for radioisotopes monitored.

- RMPs or associated documentation should address possible sample collection and storage concerns—especially those associated with progression from screening to quantitative analyses.

- Analytical laboratory methods should use EPA-approved procedures or alternatives acceptable to the state.

- Gross alpha, beta, and gamma measurements do not accurately reflect isotopic content. Comparisons with limits must be isotope specific.

The purpose of this appendix is to address the basis for the surface water RMP for the Y-12 Plant. Updates of the RMP will address specific issues associated with each plan.

With respect to the Y-12 Plant surface water program, a wide array of radioisotopes have been monitored and alpha, beta, and gamma screening measurements conducted. The monitoring history of the Y-12 Plant and changing DOE, EPA, and Tennessee requirements have resulted in procedure changes over the years and to changes in radioisotopes and sites monitored. Documentation of these changes is not readily available. Monitoring goals, which determine required detection limits based on regulations, are usually not adequate to support risk assessment for environmental restoration projects.

The lack of readily available, documented analysis that supports current monitoring leads to a need for a comprehensive overview of all monitoring data based on an overall methodology for radiological analysis that would help to identify caveats associated with existing data and provide a framework for future monitoring.

The overall radiological analysis methodology must incorporate plant operational history, results from other sites, recommendations of expert groups, and analyze whether monitoring results are consistent with physical and chemical expectations. For example, the relationships among isotopes in a decay chain are predictable if no unusual sources of a particular isotope in the chain exist, the time history is known, and differential removal or concentration mechanisms for a particular isotope are understood or do not exist. Similarly, background concentrations for radioisotopes (e.g., natural uranium and thorium series) should be consistent with those obtained in other studies given mineralogical and hydrogeological conditions similar to those at theY-12 Plant.

Radiological analysis cannot be based on monitoring results alone because monitoring at low levels against a variable background and interfering isotopes produces uncertain results. The issue of whether all isotopes are monitored that should be monitored must draw on information in addition to monitoring data.

If the radiological analysis results in lack of agreement between monitoring results and expectations based on known relationships, plant operations and transport parameters, then results are investigated further. If no explanation can be found for the unexpected results, they are suspect unless they can be confirmed by repetition. Confirmed results not in agreement with reasonable expectations stimulate investigation of the entire sample collection and analytical laboratory system.

A radiological analysis methodology has already been applied to the Y-12 Plant groundwater and surface water data for selected results and time periods. The analysis has led to an explanation of some unusual results, recommendations for future monitoring and different methods for some radioisotopes currently monitored. Specific examples are discussed in the RMPs. The analysis should be expanded to include all alpha, beta, and gamma emitters that could have been released from the Y-12 Plant over operational history.

This appendix provides an overview summary of the process. Over time, the analysis needs to be supported with more in-depth analysis and detail.

2. IDENTIFICATION OF RADIOISOTOPES

The selection of radioisotopes to be monitored in surface water is based on Y-12 Plant operational history, including waste management practices and review of groundwater, surface water, soil, sediment, settleable solids, and storm water data. Sources of radioisotopes in Y-12 Plant surface water include those in treated process liquid wastes, runoff from contaminated surface areas and groundwater which becomes surface water.

Radioisotopes that were formerly placed in waste disposal sites or were produced in the waste as a result of radioactive decay and have half-lives of more than a few years may still be detected in groundwater and surface water even though Y-12 Plant processes no longer contain these isotopes. Review of groundwater data helps to determine radioisotopes that should be monitored in surface water.

If the concentration of isotopes in groundwater are sufficiently low and no current process sources exist, then monitoring in surface water is not necessary.

Some Y-12 Plant activities involved storing low-level contaminated equipment and other materials on the surface. Spills and leaks have also occurred. Surface and subsurface soils and potentially groundwater were contaminated. Thus, surface water could receive contaminants via both surface runoff and groundwater.

A listing of radioisotopes involved in process operations at the Y-12 Plant would be all inclusive except for wastes that were transported to the Y-12 Plant from other sites. Activities at the Y-12 Plant have primarily involved uranium processing at various levels of enrichment in ^{235}U. Some recycle uranium from reactor fuel elements containing fission products and transuranics was processed from the early 1960s to 1988. Some Oak Ridge National Laboratory (ORNL) activities at the Y-12 Plant also involved fission product radioisotopes. ORNL also conducted biological research that utilized low levels of a few fission products, transuranics, and accelerator-produced tracers. Some thorium was processed at the Y-12 Plant, but thorium cycle reactor fuel elements were not processed.

Radioisotopes that were associated with these activities and could be monitored along with pertinent summaries of monitoring results through 1994 are discussed in the remainder of Section 2.

Details on the monitoring data are not accumulated here, but data continue to be collected and are being evaluated for risk assessment purposes by the Y-12 Environmental Restoration Program. Data are also reported in the ASERs, in NPDES reports, and in Groundwater Quality Assessment Reports.

2.1 URANIUM PROCESSING

Since ^{230}Th and ^{226}Ra in the ^{238}U decay series, along with corresponding radioisotopes in the ^{235}U series, were removed in the milling component of the uranium fuel cycle, the ^{238}U, ^{235}U, and ^{234}U isotopes and relatively short-lived daughters are the primary radioisotopes of monitoring interest. The ^{232}Th chain was also removed during milling. The ^{238}U daughter products , ^{234}Th and ^{234}Pa^{m}, occur up to concentrations equal to ^{238}U because secular equilibrium will be established in months. The concentrations of ^{234}Th and ^{234}Pa^{m} will not exceed ^{238}U concentrations unless there is some differential concentration mechanism for ^{234}Th and ^{234}Pa^{m} or dilution mechanism for ^{238}U. Plant processes or liquid waste treatment systems could remove uranium isotopes and leave higher concentrations of ^{234}Th and ^{234}Pa^{m} in liquid wastes. However, since these radioisotopes decay with a half-life of about 24 days, higher concentrations of ^{234}Th and ^{234}Pa^{m} unsupported by ^{238}U should have substantially decayed away unless there is a current source. Uranium may be more soluble than thorium, but the result would be higher concentrations of uranium than thorium in water.

Uranium enriched in ^{235}U by the gaseous diffusion process will have a predictable amount of ^{234}U and ^{238}U. If monitoring results are in agreement with predictions, then other processes such as those discussed above for ^{234}Th and ^{234}Pa^{m} are not important. For this reason and because enriched uranium has a higher specific activity than natural uranium, it is very important to determine the percent enrichment in ^{235}U.

Uranium analysis can be reliably based on total uranium analysis on a mass basis combined with a separate determination of ^{235}U in order to determine percent mass enrichment of ^{235}U. These determinations will allow calculation of the specific activity of total uranium and an estimate of the isotopic composition of total uranium if no other significant source of ^{234}U exists. When total uranium is less than 1 ppb, the sensitivity for determining the percent of ^{235}U will need to be improved because the specific activity for total uranium at 1 ppb mass concentration can vary from 0.67 pCi/μg for natural uranium or less for depleted U to 67 pCi/μg for 95 percent enrichment.

In the past, total uranium estimates based on activity concentrations of ^{238}U, ^{235}U, and ^{234}U determined by alpha spectrometry have been significantly higher than total uranium results based on mass spectrometry. The difference is most likely due to contributions of other natural and transuranic alpha emitters to the energy peaks analyzed for ^{234}U, ^{235}U, and ^{238}U. Matrix effects in the samples and in the sample holders could produce energy straggling, which could significantly reduce resolution. These possibilities are especially important when concentrations approach background concentrations.

The analytical laboratory radiochemically separates the analyte and spikes real-world samples so that system response for a known added amount of the analyte can be determined. This practice is quantitative for individual radioisotopes but in mixtures of radioisotopes only allows quantitative results if interfering isotopes can be accounted for.

Monitoring results to date indicate that most uranium in water samples is slightly depleted (^{235}U less than 0.72 weight percent) so that measurement of total uranium and the calculation of isotopic composition assuming natural uranium would overestimate the concentrations of ^{235}U and ^{234}U and underestimate the concentration of ^{238}U. However, there are enough exceptions where the uranium appears to be enriched so that total uranium analysis alone is not sufficient to determine isotopic activities. If the calculation of total uranium specific activity is based on ^{234}U, then the detection limit requirement could be substantially relaxed. The concentration of ^{234}U for 1 ppb natural U is about 0.33 pCi/L. The concentration of ^{234}U in total uranium at 1 ppb enriched to 95 percent in ^{235}U is about 65 pCi/L. For natural uranium, the concentration of ^{235}U at 1 ppb total uranium would be 0.014 pCi/L. For 95 percent enriched uranium the ^{235}U concentration is 0.95 ppb or 1.9 pCi/L. The approach would determine total uranium by mass spectrometry and ^{234}U by alpha spectrometry.

The isotopic compositions at various levels of enrichment in ^{235}U for a total uranium concentration of 1 ppb are given in Table 1. For other levels of total uranium, each column in Table 1 except the first column would be multiplied by the total uranium content in ppb. A ^{234}U detection limit of 0.1 pCi/L is adequate to fully characterize uranium isotopes based on total uranium and ^{234}U analysis.

The calculations are as follows after measurements of total U (ppb) and ^{234}U (pCi/L).

1. Use Table 1 to determine weight percent ^{235}U from ^{234}U measurement.

2. Calculate specific activity (SPA) of total uranium from Rich formula (Rich et al., 1988).

$$\text{SPA total U} = 0.4 + 0.38\,(\%\,^{235}U) + 0.0034\,(\%\,^{235}U)^2\ \text{pCi}/\mu\text{g}.$$

$$\text{Total U (pCi/L)} = \text{total U (ppb)} \times \text{SPA total U (pCi}/\mu\text{g)}.$$

At this point results could be compared with concentration limits for total U. If values for the specific isotopes are required, then the following relationship can be used

$$(^{238}U + {}^{234}U)\ \text{pCi/L} = \text{total U (pCi/L)} - {}^{235}U\ \text{(pCi/L)}.$$

Since ^{234}U and total U are known and ^{235}U can be determined from percent enrichment of ^{235}U, ^{238}U can be calculated.

The required measurements are total uranium with a detection limit of 1 ppb and ^{234}U with a detection limit of 0.1 pCi/L for depleted uranium. Expected equilibrium of ^{234}Th and $^{234}Pa^m$ with ^{238}U provides for their estimation. Possible interferences with ^{234}U measurements must be removed by radiochemical techniques for accurate analysis. Possible interfering isotopes are discussed further for transuranium isotopes in Section 2.2. Note that such low detection limits are required for uranium measurement at or near background concentrations. Substantially higher detection limits may be adequate for regulatory monitoring (See Section 3 of this appendix).

Table 1. Isotopic Composition of a 1 ppb Total U Water Sample versus Weight Percent of ^{235}U

Weight % ^{235}U	^{235}U (ppb)	Total U (pCi/L)	^{238}U (pCi/L)	^{234}U (pCi/L)	^{235}U (pCi/L)
0.1	0.001	0.438	0.327	0.0109	0.002
0.2	0.002	0.476	0.326	0.146	0.004
0.3	0.003	0.514	0.326	0.182	0.006
0.4	0.004	0.552	0.326	0.218	0.008
0.5	0.005	0.591	0.325	0.256	0.010
0.6	0.006	0.629	0.325	0.292	0.012
0.7	0.007	0.668	0.328	0.329	0.014
1	0.01	0.78	0.324	0.438	0.02
2	0.02	1.12	0.320	0.814	0.04
5	0.05	2.38	0.311	1.974	0.10
10	0.1	4.54	0.294	4.046	0.20
20	0.2	9.36	0.262	8.698	0.40
30	0.3	14.86	0.229	14.031	0.60
40	0.4	21.04	0.196	20.044	0.80
50	0.5	27.90	0.163	26.737	1.00
60	0.6	35.44	0.131	34.109	1.20
70	0.7	43.66	0.098	42.162	1.40
80	0.8	52.16	0.065	50.495	1.60
90	0.9	62.14	0.033	60.307	1.80
95	0.95	67.18	0.0163	65.269	1.90
100	1	72.40	0	70.40	2.00

Early monitoring results for Y-12 Plant surface water included only total uranium and weight percent ^{235}U. These results were adequate given the assumption that ^{234}Th and ^{234}Pa^{m} are in equilibrium with ^{238}U. More recent measurements have included ^{234}Th and ^{234}Pa^{m,} with results sometimes indicating ^{234}Th and ^{234}Pa^{m} at levels higher than ^{238}U. However, the elevated ^{234}Th and ^{234}Pa^{m} results relative to ^{238}U are suspect unless there is a current source for ^{234}Th and ^{234}Pa^{m}. Overall, the bulk of the measurements for surface water and groundwater indicate that uranium in the aquatic system at the Y-12 Plant is depleted. So ^{234}U concentrations are generally less than ^{238}U concentrations. Since the half-life of ^{234}U is very long compared with its parents except for ^{238}U, the ^{234}U present is due to decay from ^{238}U or from enrichment.

The early measurements also did not include fission products and transuranics which became important after recycle uranium processing began in the early 1960s. The basis for the fission product and transuranic monitoring program is discussed in Section 2.2.

2.2 RECYCLE URANIUM PROCESSING

Recycle uranium from reactor fuel elements was processed at the Y-12 Plant from the early 1960s to April 1988. Fission products and transuranic isotopes occur as a result of these activities. Since no current process source exists, radioisotope concentrations in groundwater associated with waste sites will generally represent maximum potential concentrations in surface water. Direct contact of surface water with waste masses resulting in runoff to surface water does not occur.

Small amounts of recycle uranium were processed relative to the amounts from enrichment processes. Fission products and activation products were low in recycled uranium relative to the amount in fuel elements because they were reduced during processing of the fuel elements at other sites. Transuranics were also associated with recycle uranium. Wastes from processing recycle uranium were placed in the S-3 Pond. Therefore, groundwater monitoring results for the S-3 Pond include contributions of fission products and transuranics from recycle uranium processing.

A qualitative conservative analysis of fission products is presented because details of reactor type, operation, and fuel element processing will require further study. However, a qualitative analysis, assuming direct fuel element processing, provides a comprehensive basis for identifying isotopes that could be monitored. Knowledge that the reactor was fueled by ^{235}U is enough to approximate roughly the relative amounts of fission products produced per gram of ^{235}U fuel consumed. Analysis of monitoring data and general experience provides a basis for testing preliminary qualitative plans. Yield values for the fission products produced by thermal neutron bombardment of ^{235}U may be taken from the familiar mass yield curve given in numerous textbooks and in the *Radiological Health Handbook* (1992). More than 60 primary products have been identified. These products are all radioactive and produce, on average, about three other radioactive isotopes before a stable species is formed. Many of the isotopes have very short half-lives and/or are produced in low yields so that in a few years only a subset of the possible isotopes contribute the bulk of the activity.

A list of isotopes contributing an arbitrary amount of more than 1 mCi per gram of ^{235}U consumed is given in Table 2. The values in Table 2 apply for 30 years after removal from a reactor. This is only an example calculation. Shorter times after processing would yield different relative values for some of the isotopes. The implications for the isotopes to be monitored in the future based on the analysis that follows remain qualitatively valid. The contributions from the decay of other isotopes has been added to the amounts calculated from the yields given in Table 1. For example, to the amount calculated from the fission yield for ^{137}La is added ^{137}La from the decay of ^{137}Ce and ^{137}Pr, which are produced at about the same fission yield as ^{137}La.

The activity values listed for the isotopes in Table 2 are only approximate and not all inclusive. For example ^{239}Pu produced via the reaction

$$^{238}U(n,\gamma)^{239}U \xrightarrow[23\text{ min}]{} {}^{239}Np \xrightarrow[2.3\text{d}]{} \beta\,{}^{239}Pu$$

will fission and produce a slightly different mass yield curve than for ^{235}U fission. However, the cross section for the $^{238}U(n,\gamma)^{239}U$ reaction is only about 2.8 barns while the cross section for fission of ^{235}U is about 550 barns. Thus, for reactors enriched in ^{235}U, the production of ^{239}Pu fission products is expected to be much lower that for ^{235}U. However, even if the reactor wre fueled with ^{239}Pu, fission product yields would not differ substantially from those for ^{235}U. Transuranium elements of higher mass— ^{240}Pu, ^{241}Pu, and ^{242}Pu will be produced. ^{241}Pu will fission and is also the first member of the neptunium series which includes ^{241}Am, ^{237}Np, ^{233}Pa, etc. The tranuranic elements are discussed further below.

Also listed in Table 2 are the principle radiation emissions of the fission products. It is readily apparent that for non-isotope-specific beta counting methods like liquid scintillation counting or gas flow proportional counting, many isotopes can contribute to the total beta decay. Radiochemical preparation methods must be very specific to a particular isotope in order to obtain quantitative results for that isotope. Given the relative amounts of the isotopes listed in Table 2, 99 percent separation will not rule out potential significant interferences among isotopes.

The fact that background concentrations have been reported for fission products when concentrations should actually be very low is evidence for contributions from natural beta-emitting radionuclides to the energy ranges analyzed for specific isotopes.

The overall result of these possible interferences is that the concentration of each isotope will be overestimated. Therefore, if the isotopes selected for monitoring are the ones with lowest regulation or guidance concentrations (e.g., ^{90}Sr among the fission products), then the overall result could be extremely

Table 2. Fission Products Contributing More than 1mCi per Gram of ^{235}U Consumed 30 years After Removal From a Reactor

Isotope	Yield (5) Approx.	Half-Life (years)	Activity per gram ^{235}U (Ci)	Emmission	Energy	(MeV)
				α	β	γ
Kr-85	1	10.72	0.56	0	0.251	0.002
Nb-93m	6	13.6	3.62	0	0.028	0.002
Sr-90	6	29.12	3.96	0	0.196	0
Y-90	Secular	equilibrium	with Sr-90			
Nb-94	6	2.03E+04	1.11E-02	0	0.168	1.574
Mo-93	6	3.50E+03	4.50E-02	0	0.006	0.011
Tc-99	6	2.13E+05	1.50E-03	0	0.101	0
Rh-101	5	3.2	0.12	0	0.032	0.069
Rh-102	4	2.9	3.50E-02	0	0.012	2.140
Ag-108m	0.02	127	4.37E-03	0	0.016	1.627
Ag-108	Secular	equilibrium	with	Ag108m		
Cd-113m	0.01	13.6	5.00E-03	0	0.185	0
Sn-121m	0.01	55	3.63E-03	0	0.035	0.005
Ba-133	5	10.74	0.88	0	0.054	0.402
La-137	6	6.00E+04	2.58E-03	0	0.007	0.024
Pm-145	4	17.7	0.85	0	0.014	0.031
Pm-146	4	5.53	0.41	0	0.097	0.753
Pm-147	3.5	2.62	0.01	0	0.02	0
Eu-150	1	34.2	0.37	0	0.044	1.496
Eu-152	0.6	13.33	0.22	0	0.139	1.155
Eu-154	0.1	8.8	0.02	0	0.292	1.242
Eu-155	0.03	4.96	1.00E-03	0	0.063	0.061
Gd-148	1	93	0.2	3.183	0	0
Tb-157	0.01	150	1.30E-03	0	0.005	0.003
Tb-158	0.01	150	1.30E-03	0	0.116	0.798

conservative. A significant fraction of the historic database may be confounded by such interferences because isotope concentrations, especially those with shorter half-lives, were higher; separation methods have improved; and counting systems have improved providing lower detection limits.

Identification of isotopes that should be monitored can be based on a combination of concentrations and toxicities. A risk based determination would involve knowledge of amount in process or waste inventory, amount released, transport to exposure points, intake at exposure points and dose per unit intake or dose conversion factors (DCFs). The site specific factors (all except DCFs) were not known in the past and still are not known quantitatively. Measurement of concentrations is necessary because the factors are not quantitatively known. However, determination of which isotopes to measure initially and whether current measurement programs are complete must be based on general (not site specific) knowledge of the technology. Decisions are based on relative properties of the isotopes rather than quantitative estimates for each one. As data accumulates, confidence in the choices will increase. The important relative properties include:

- relative amounts in the process or in wastes,
- amounts released over time (source term),
- tendency to disperse in the environment,
- persistence (represented by half life and chemical properties), and
- toxicity (represented by DCF)

For example ^{90}Sr, ^{137}Cs, and ^{3}H tend to disperse more than other fission or activation products in water. They are produced in higher quantities than the other fission or activation products. ^{90}Sr and ^{137}Cs have relatively high dose conversion factors and relatively long half lives.

Based on this general knowledge, ^{90}Sr can be selected as a reference isotope for fission and activation products to estimate relative hazard (risk potential) on a qualitative basis for selection of isotopes to monitor. Information on relative concentrations in the representative fuel elements, 30 years after removal from a reactor, relative dose conversion factors and relative hazard potential referenced to ^{90}Sr are given in Table 3. The 30 year time frame is an arbitrary choice for qualitative analysis but is perhaps appropriate for future monitoring activities. Isotopes with hazard potential less than 10^{-4} are not listed in Table 3. Based on the estimates in Table 3, if ^{90}Sr and ^{137}Cs are shown to have acceptable concentrations, then the other isotopes will have acceptable concentrations by a wide margin. This is a conservative conclusion because of the inherent assumption that all isotopes disperse as readily as ^{90}Sr. It is also assumed that all fission and activation products have the same relative concentrations in wastes as in fuel elements.

Table 3. Relative Hazards of Fission Radioisotopes in a Representative Reactor Fuel Element

Isotopes	Ci/Ci^{90}Sr(a)	Ingestion DCFi/DCF^{90}Sr	Relative Hazard Potential
H-3	?	4.50E-04	?
Co-60	1.90E-03	0.19	3.60E-04
Sr-90	1	1	1
Y-90	1	2.56E-02	2.56E-02
Nb-93m	1	3.66E-03	3.66E-03
Rh-101	3.00E-02	1.63E-03	4.90E-04
Rh-102	8.84E-03	7.32E-02	6.50E-04
Cs-137	0.65	0.35	0.23
Ba-137m	0.62	1.29E-02	8.00E-03
Ba-133	0.22	2.38E-02	5.25E-03
Pm-145	0.21	3.32E-03	6.97E-04
Pm-146	0.10	2.57E-02	2.57E-03
Pm-147	2.50E-03	7.35E-03	1.84E-05
Eu-150	9.30E-02	4.46E-02	4.15E-03
Eu-152	5.50E-02	4.54E-02	2.50E-03
Eu-154	5.00E-03	6.70E-02	3.35E-04
Gd-148	5.05E-02	1.53	7.73E-02

(a)Ci/C^{90}Sr = Curies of isotope listed divided by Curies of ^{90}Sr.

Tritium is often monitored around reactors because it is so readily dispersed. Since more tritium is produced in reactor coolants, this would not necessarily extrapolate to other situations. These indications are consistent with recommendations for monitoring around power reactors based on experience. For example the Radiological Health Handbook recommends gamma spectrometry (e.g. ^{137}Cs or Europium isotopes) ^{90}Sr and ^{3}H monitoring on quarterly composites for surface water and add ^{226}Ra on annual composites for groundwater.

Most activation products have short half lives. Contaminants which continue to contribute after a few years include ^{3}H and ^{60}Co. ^{3}H may be produced by neutron irradiation of ^{2}H, ^{3}He, ^{6}Li, ^{7}Li, ^{9}Be, ^{10}Be, ^{12}C and ^{14}N. The half life of ^{3}H (12.3 years), its mobility and amounts in reactor fuel elements indicate monitoring until concentrations are shown to be acceptably low. ^{60}Co is usually produced by neutron irradiation of ^{59}Co. The 10.3-minute isomeric state of ^{60}Co is also produced by thermal neutron bombardment of ^{59}Co. Greater than 99 percent of the decays of this isomeric state lead to the 5.27 year ground state of ^{60}Co. Relatively small amounts for ^{60}Co result continuously from the decay of 1E+05 year ^{60}Fe which decays to ^{60}Co^{m}. Since ^{60}Co is produced in the structural components of fuel elements, it is not as readily released as the fission products.

Past monitoring has included ^{60}Co, ^{90}Sr, ^{137}Cs, ^{3}H and ^{99}Tc. The inclusion of ^{3}H and ^{99}Tc is based on their high mobility and the fact that they have been identified in groundwater plumes at the Y-12 Plant. They serve as indicators of the extent of transport of fission and activation product radioisotopes.

Review of groundwater and surface water data for the Y-12 Plant indicates that the concentrations of fission products are generally low relative to the uranium isotopes. The only isotopes that exceed 4 mrem/yr drinking water criteria in the most contaminated on-site wells near the S-3 Site are ^{99}Tc and ^{90}Sr. Concentrations of ^{90}Sr are less than half the 1000 pCi/L DCG for the most contaminated on-site well. At Stations 17 and 304 which represent Y-12 Plant off-site surface water releases ^{90}Sr concentrations are about 0.1 - 0.2 percent of the DCG (about 2.5 - 5 percent of drinking water criteria).

The indication that ^{90}Sr may be the key fission product radioisotope based on qualitative analysis is supported by the monitoring data. Although present only at very low levels in surface water, it is present in groundwater which is a surface water source. Therefore, monitoring should continue with further study of the possibility of interferences by other beta emitters.

The analysis of direct fuel element processing, as a conservative surrogate for recycle uranium processing, is certainly incomplete without consideration of the transuranic radioisotopes that are formed in reactors. Many of these radioisotopes are more toxic than ^{90}Sr and have very long half lives. Thus, they can represent a long term hazard much greater than ^{90}Sr if present in sufficient quantity. Estimates of transuranic formation from the reactor safety study are given in the Radiological Health Handbook (1992). These estimates may be used for a qualitative estimate of the relative hazard potential of the transuranics similar to the estimates given in Table 3 for the fission products. One such set of estimates is given in Table 4.

Some progeny of transuranics may be important because of their amount and toxicity but also because they may interfere with analyses for other isotopes. The isotope ^{237}Np, the daughter product of

^{241}Am can also be produced via the reaction:

$$^{236}\mathrm{U}\ (n,\gamma)\ ^{237}\mathrm{U} \xrightarrow[6.75\mathrm{d}]{\beta^-} {}^{237}\mathrm{Np} \xrightarrow[2.14 \times 10^{6}\mathrm{yr}]{\alpha} {}^{233}\mathrm{Pa}.$$

To a lesser extent ^{237}U will be produced by ^{238}U (n, 2n) and ^{235}U (2n,g) reactions. Concentrations of ^{237}Np could be higher than ^{241}Am in spite of the fact that it has a half life much greater than its parent. ^{237}Np emits an alpha (4.769 MeV) only 11 keV above ^{234}U (4.758) and decays to ^{233}Pa which emits a beta spectrum of weighted average energy exactly the same as ^{90}Sr (0.195 MeV). ^{233}U, the daughter of ^{233}Pa, emits alphas of 4.78 MeV (13.2 percent), only 2 keV from ^{234}U and 4.82 MeV (84.4 percent) about 60 keV above ^{234}U and 50 keV above ^{237}Np. ^{226}Ra emits an alpha (4.774) which is only 5 keV above ^{237}Np and 16 keV above ^{234}U. With typical alpha spectrometry full width half maximum resolutions of 50-75 keV, the radiochemistry will have to be very selective in order to discriminate among these isotopes at low concentrations. The uranium/radium separation may be very high while equivalent neptunium/uranium separation may be more difficult because of similar chemistry.

Table 4. Relative Hazard of Transuranics in a Representative Fuel Element

Isotopes	Ci/Ci^{90}Sr	Ingestion DCFi/DCF^{90}Sr	Relative Hazard Potential
Sr-90	1	1	1
Pu-238	2.20E-02	22.5	0.5
Pu-239	8.00E-03	24.8	0.2
Pu-240	8.00E-03	24.8	0.2
Pu-241	0.31	0.48	0.15
Am-241	6.54E-04	25.5	1.[illegible]E-02
Cm-244	2.81E-03	14.1	4.0[illegible]E-02

As was the case for beta emitters, interferences would lead to overestimates of transuranic concentrations. Comparisons of measured concentrations to concentrations limits will be conservative.

The transuranium radioisotopes are expected to be less mobile than ^{90}Sr, ^{137}Cs, ^{3}H or ^{99}Tc.

Therefore, although their hazard potential may be comparable to ^{90}Sr and exceed ^{90}Sr after another 30 years, the risk associated with the transuranics may be very low. Measurements for Pu isotopes for the most contaminated on-site wells associated with the S-3 Pond site indicate that they are either not mobile and/or are present in the wastes at low levels. Monitoring in groundwater (source) should continue. Concentrations of transuranics in surface water presently are low enough so that they are not of concern from NPDES regulatory perspective (See Section 3).

2.3 THORIUM PROCESSING

Thorium processing at the Y-12 Plant involves considerations similar to uranium processing. If ^{232}Th and ^{228}Th are selectively removed in a Y-12 Plant process, then ^{228}Ra and ^{224}Ra could be elevated in liquid wastes. However, in a time frame of 5–10 years ^{228}Th should achieve equilibrium with ^{228}Ra. Decay of ^{228}Ra plus turnover in groundwater and/or surface water should have significantly diluted initial concentrations. Since ^{232}Th is not currently processed at the Y-12 Plant, any current releases are from wastes sites through groundwater. Groundwater data (1993, 1994) do not show ^{228}Ra sufficiently elevated to account for surface water levels measured in 1991. Later surface water measurements (1993–1994) for ^{228}Ra are much lower than 1991 levels and are much more in line with expectations. Results were discussed in Section 5.2 of the previous version of the RMP.

Monitoring for these natural isotopes ^{232}Th, ^{228}Ra, ^{228}Th to determine their background concentrations and their contributions to the integrated dose limit will require lower MDAs than practical for routine monitoring (see Section 3). Other natural radionuclides in the ^{238}U decay chain, specifically ^{226}Ra and ^{230}Th, should be included in special monitoring activities because they can interfere with measurements for other isotopes and can make a significant contribution to the integrated dose limit.

3. BASIS OF DETECTION LIMITS

Analytical systems should have sufficient accuracy and precision to measure concentrations at monitoring goals based on regulatory limits, background levels or for other uses such as risk assessment, ALARA studies, etc. Monitoring background levels or for other uses is not currently practical for a regulatory activity. DOE has provided derived concentration guides (DCGs) based on a 100-mrem/yr reference dose assuming direct intake or exposure. However, the ALARA philosophy requires that actual doses be as far below reference doses as reasonably achievable. Also in the proposed codification of DOE Orders 5400.1 and 5400.5, the total effective dose equivalent from all isotopes from all sources and through all pathways is limited to 100 mrem/yr for a member of the public. Therefore, the dose through one pathway and/or from one source must usually be less than 100 mrem/yr. The corresponding concentration limit and associated measurement capability must therefore be only a fraction of DGGs. The detection limit for each isotope must be even lower since all isotopes are to be compared with their limits and summed to determine compliance. That is

$$\Sigma \mathrm{Ci}/\mathrm{CLi} < \;,$$

where Ci is the measured concentration of isotope i and CLi is the limiting concentration of isotope i at the exposure point. As a simplistic example, given n isotopes with limiting concentrations CLi, the DL for each isotope would need to be less than CLi/n.

If concentrations are not measured at exposure points, then the limiting concentrations must be extrapolated (modeled) to the measurement point or the measured concentration must be extrapolated to the exposure point. This extrapolation is accomplished by a pathways analysis which we indicate here as a transport factor (TF). Using TF as a simple volume dilution factor for surface water is sufficient for our purposes. Ignoring other important processes (e.g., sedimentation) is conservative for the drinking water pathway. These TFs are site dependent making the DLs also site dependent.

In either case, the summation becomes

$$\Sigma \mathrm{CiTF}/\mathrm{CLi} < 1.$$

In this case the MDA for each of n isotopes would have to be CLi/TFn. For surface water releases diluted by the Clinch River from the Y-12 Plant, the TF is about 0.001 (Bear Creek or East Fork Poplar Creek Flow divided by Clinch River or Tennessee River flow at a potential drinking water exposure point.) In this case the relevant reference dose is taken to be the limit of 4 mrem/yr for the drinking water pathway integrated over all isotopes and all sources. This is not a regulatory limit, but it would ensure that regulatory limits are not exceeded for any isotope.

Under current conditions, MDAs for measurement at Y-12 Plant stations to surface water need only to be about 1000CLi/n, where CLi is based on the 4-mrem/yr dose or about 40 DCG/n. If 20 isotopes are monitored and allowing for contribution of other sources (three major ones for ORR) which may make a larger contribution than the Y-12 Plant, and pathways (e.g., groundwater to Clinch River, unmonitored surface water or aquatic food chain), MDAs at about 0.2 DCG concentrations appear adequate to quantify the Y-12 Plant surface water component of reference doses (4-mrem/yr integrated dose). However, such MDAs are not adequate to quantify isotopes that occur at lower levels.

For the DOE Part 834 reporting requirement of 10 mrem/yr, with water and air pathway considered independently, the MDAs for Y-12 Plant surface water would need to be about 0.5 DGGs.

The analytical laboratories processing water samples from the Y-12 Plant currently have the capability to analyze at detection limits well below those required for regulatory analysis. Lower detection limits will be necessary because of the interests of various user groups, not because of regulatory needs. These user groups include those interested in defining contamination plumes (hydrogeological transport studies) with respect to background conditions; ALARA studies; risk-assessment studies, which generally require data on background conditions and which seek to quantify risks at levels well below those associated with regulatory levels at future times when people may have direct access to the site (transport factor = 1). Isotopes well below even drinking water concentrations continue to be monitored (e.g., tritium, technetium) because they are perceived to generate public interest, serve as tracers, or are present in groundwater.

Measurement at low levels given a highly complex and variable background presents significant problems associated with interferences and can easily produce false positive results. The level of effort required to separate isotopes and convert them to a form suitable for accurate and sensitive analysis is much greater than that required when isotope concentrations are significantly above background.

Typical MDAs currently reported by the Analytical Services Organization are listed in Table 5 along with sample volumes, counting times, and counting method. These MDAs are the lowest achievable without significant escalation in time and cost which are not justified for a routine regulatory program.

The MDAs in Table 5 represent practical monitoring goals for NPDES requirements. Isotopes with concentrations below these MDAs could be confidently eliminated from future NPDES monitoring.

Determination of background concentrations of some natural radionuclides in water will require even lower MDAs than given in Table 5. Special studies would be required to determine background concentrations. According to an EPA summary given in the September 30, 1986, draft of the advanced notice of proposed rulemaking for radionuclides in drinking water (40 CFR Part 141), the population weighted average of natural radionuclides in the U.S. community drinking water is:

Ra-226	0.3 - 0.8
Ra-228	0.4 - 1.0
Total U	0.3-2.0
Th-230	<0.04
Th-232	<0.01

Table 5. **Typical MDAs versus Sample Volumes and Counting Times**

Isotope	Typical Sample Size (L)	Typical Count Time (hours)	Typical MDA* (pCi/L)	Method
U-238	0.1	16.7	0.5	Alpha spectrometry
U-235	0.1	16.7	0.5	Alpha spectrometry
U-234	0.1	16.7	0.5	Alpha spectrometry
Am-241	0.1	16.7	0.5	Alpha spectrometry
Np-237	0.1	16.7	0.8	Alpha spectrometry
Pu-238	0.1	16.7	0.5	Alpha spectrometry
Ra-226	0.5	1	1	Gas flow prop.
Ra-228	0.5	1	3	Gas flow prop.
Th-228	0.1	16.7	0.5	Alpha spectrometry
Th-230	0.1	16.7	0.5	Alpha spectrometry
Th-232	0.1	16.7	0.5	Alpha spectrometry
H-3	0.05	1	400	Liquid scintillation
Tc-99	0.4	1	8	Liquid scintillation
Sr-90	0.2	1	8	Gas flow prop.
Cs-137	0.9	16.7	6	Gamma spectrometry

*The MDAs also depend on counter efficiency and recovery which will vary with sample matrix.

The values represent the average of surface water and groundwater supplies in pCi/L. The values for ^{230}Th and ^{232}Th are below MDAs for the usual analytical procedures and are well below levels that would make a significant contribution to the integrated dose.

The values for total uranium are for natural uranium which at 1 pCi/L would contain about 0.33 pCi/L of ^{238}U, 0.33 pCi/L of ^{234}U and about 0.014 pCi/L of ^{235}U. Therefore, the MDAs necessary to determine background concentrations of natural uranium are lower than MDAs in Table 5. Similar to the thorium background values, the uranium isotopes at background concentrations will not make a significant contribution to the integrated dose.

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