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July 25, 1995

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Mr. R. J. Spence  
Department of Energy, Oak Ridge Operations  
Post Office Box 2001  
Oak Ridge, Tennessee 37831

Dear Mr. Spence:

**Radiological Monitoring Plan for the Oak Ridge Y-12 Plant: Surface Water**

National Pollutant Discharge Elimination System (NPDES) Permit TN0002968, issued April 28, 1995, requires that the Y-12 Plant Radiological Monitoring Plan for surface water be modified (Part III-H). These modifications shall consist of expanding the plan to include storm water monitoring and an assessment of alpha, beta, and gamma emitters. In addition, a meeting was held with personnel from the Tennessee Department of Environment and Conservation (TDEC) on May 4, 1995. In this meeting, TDEC personnel provided guidance to Y-12 Plant personnel in regard to the contents of the modified plan.

Enclosed is a revised plan incorporating the permit requirements and guidance provided by TDEC personnel. In addition, modifications were made to address future requirements of the new regulation for radiation protection of the public and the environment (Title 10, Code of Federal Regulations, Part 834) in regards to surface water monitoring.

Please forward this plan by August 1, 1995, to:

Mr. Larry Bunting  
Tennessee Department of Environment and Conservation  
Division of Water Pollution Control  
401 Church St., 6th Floor, L&C Annex  
Nashville, Tennessee 37243-1534

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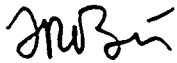
Mr. R. J. Spence, DOE-ORO

Page 2

July 25, 1995

If further explanation or discussion of this plan is desired, please contact L. O. Vaughan at 6-8108.

Very truly yours,



T. R. Butz  
Y-12 Plant Manager

TRB:rf

Enclosure: As Stated

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Enclosure  
Letter, Butz to Spence  
Dated: July 25, 1995

LETTER TITLE

**Radiological Monitoring Plan for the Oak Ridge Y-12 Plant: Surface Water**

# Y-12

**OAK RIDGE  
Y-12  
PLANT**

**RADIOLOGICAL MONITORING PLAN  
FOR  
THE OAK RIDGE Y-12 PLANT: SURFACE WATER**

**MARTIN MARIETTA**

**JULY 1995**

**Prepared by**

**H&R TECHNICAL ASSOCIATES, INC.  
UNDER PURCHASE ORDER 90Y-TK532C**

**for the**

**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 No. DE-AC05-84OR21400**

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

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**RADIOLOGICAL MONITORING PLAN**  
**FOR**  
**THE OAK RIDGE Y-12 PLANT: SURFACE WATER**

**Environmental Management Department**  
**Health, Safety, Environment and Accountability Division**

Oak Ridge Y-12 Plant  
Oak Ridge, Tennessee 37831

Operated by Lockheed Martin Energy Systems  
for the U.S. Department of Energy  
Under Contract No. DE-AC05-84OR21400

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## ACRONYMS

ALARA	As low as reasonably achievable
ANPR	Advance Notice of Proposed Rulemaking
ANSI	American National Standards Institute
ASER	Annual Site Environmental Report
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
ELIMS	Environmental Laboratory Management Information 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
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
TDEC	Tennessee Department of Environment and Conservation
TEDE	Total effective dose equivalent

## 1.0 INTRODUCTION AND BACKGROUND

### 1.1 PURPOSE

The Y-12 Plant in Oak Ridge, Tennessee, is operated by Lockheed Martin Energy Systems (Energy Systems) for the United States Department of Energy (DOE). Therefore, the Y-12 Plant has been operated 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's 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.

The state of Tennessee Division of Water Pollution Control has recently communicated their needs and goals in addition to NPDES requirements for 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 has been prepared to provide an overview of radiological assessment that begins to address these current needs and goals. Appendix A is intended to supplement documentation provided in the Annual Site Environmental Reports (ASERs), NPDES reports, Groundwater Quality Assessment Reports and studies conducted by the Y-12 Plant Environmental Restoration (ER) Program to provide a more complete documented basis for the current Y-12 Plant surface water monitoring program. The goal of this supplemental effort to the specific RMPs is to eventually provide a comprehensive overview that documents the basis of the Y-12 Plant surface water monitoring program and an acceptable analysis methodology for directing future monitoring.

The purpose of this update to the Y-12 Plant RMP is to satisfy the requirements of the current NPDES permit and 10 CFR Part 834 as presently anticipated by defining the radiological monitoring plan for surface water for the Y-12 Plant. This plan includes initial storm water monitoring and data analysis. Monitoring of the sanitary sewer system is not included in this version of the plan.

The plan discusses the requirements of pertinent DOE orders particularly 5400.5 as codified via 10 CFR Part 834 (proposed) with respect to design goals for monitoring sensitivity (monitoring goals) necessary to determine background concentrations of radionuclides, to quantify releases to determine trends, to satisfy regulatory requirements, to support consequence assessments, and to meet requirements that releases be "as low as reasonably achievable" (ALARA).

The RMP has evolved from a plan to meet only NPDES requirements to a plan that considers other monitoring goals to meet the needs of other users to the extent practical. The monitoring goals for other users are not requirements of the NPDES RMP but to the extent that they can be accommodated at reasonable costs, less redundancy and lower long term costs will result.

The requirements of DOE Order 5400.5 as will be codified in 10 CFR Part 834 must be met for the Y-12 Plant surface water monitoring program. The requirements of Part 834 will also met the requirements of the NPDES Permit. These requirements are summarized below in order to identify monitoring goals to meet present and anticipated regulatory requirements.

Monitoring goals to meet the requirements of consequence analysis or determination of background are generally below those associated with NPDES requirements. Additional effort and costs are associated with achieving detection limits to meet these monitoring goals. These goals are most efficiently accomplished by an extension of the ongoing program to meet regulatory requirements.

ALARA considerations are discussed in the context of achieving goals as far below regulatory requirements as practical. Actual ALARA goals based on a detailed ALARA plan are not documented in this RMP. Actual ALARA goals are not likely to be lower than practical monitoring goals identified in this RMP which are well below monitoring goals based on regulatory requirements. Lower ALARA goals for a regulatory monitoring program would involve significant escalation in effort and costs.

These aspects of the plan are included not for formal approval by TDEC Clean Water Program staff but to insure a cohesive and complete discussion of the subject. Regulatory authority for requirements of 10 CFR 834 and for DOE orders lies with the DOE.

## 1.2 BACKGROUND

The roles and requirements for the agencies involved in regulations, control and guidance for the Y-12 Plant water monitoring program are summarized below. The state of Tennessee Division of Water Pollution Control goals and requirements are discussed in more detail in Appendix A. In addition, surface water monitoring is part of the responsibility of the DOE Oversight Division of the Tennessee Department of Environment and Conservation (TDEC).

The most current DOE guidance is contained in the January 1991 report titled *Environmental Regulatory Guide for Radiological Effluent Monitoring and Environmental Surveillance* (DOE/EH-01735). This regulatory guide presents elements of a radiological effluent monitoring and environmental surveillance program considered acceptable to DOE in support of DOE 5400.5 (Radiation Protection of the Public and the Environment) and DOE 5400.1 (General Environmental Protection Program).

The DOE has proposed to codify the requirements of DOE Order 5400.5 into the Code of Federal Regulations as 10 CFR Part 834 (Part 834). The codification will strengthen the order by establishing enforcement procedures and penalties for non-compliance.

Part 834 incorporates the philosophy and recommendations of Publication 26 of the International Commission on Radiological Protection (ICRP) and Report No. 91 of the National Committee on Radiological Protection (NCRP) as do other major federal standards developed by the Nuclear Regulatory Commission (NRC) and the Environmental Protection Agency (EPA).

Part 834 will require an Environmental Radiation Protection Plan (ERPP) for every activity operated by the DOE. The level of effort (degree of detail) required in an ERPP should be commensurate with the risk potential of the activity. Activities involving small risks should require minimal efforts to prepare an ERPP. An ERPP produced for one activity may also be applicable for other similar activities. The ERPP may incorporate other plans, documents or sections of other plans and documents. The ERPP may be incorporated into another plan such as a site environmental monitoring plan.

Essentially all the requirements in Part 834 were required in DOE Order 5400.5. Since the current NPDES Y-12 Plant RMP considers the requirements of DOE Order 5400.5, the update 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, any 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.

The DOE regulatory guide is expected to be revised to correspond to Part 834. Currently, the guide denotes high priority procedures with the word "should" followed by an asterisk (should\*). Guidance procedures 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 most pertinent general 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 ( $1.95 \sigma$  where  $\sigma$  is the standard deviation of the net counting rate of the sample). . . ." It should be noted here that the Y-12 Plant effluents and surface water streams are not drinking water supplies and not subject to the requirements of the Safe Drinking Water Act.

The most recent U.S. Environmental Protection Agency (EPA) proposed regulations on radionuclides in drinking water are contained in an Advance Notice of Proposed Rulemaking (ANPR) entitled *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 for NPDES outfalls.

Taken together, the DOE high priorities and 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 to 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), to 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 obvious. However, DOE has established derived concentration guides (DCGs) equivalent to 100 mrem/yr by each pathway for all radioisotopes, and the state requires that measured concentrations be compared to the DCGs.

Part 834 requires that operation be conducted to ensure that doses to members of the general public and releases of radioactive material into the environment:

- do not exceed established dose limits or DOE release limits.
- are kept ALARA, and
- conform the restrictions and conditions in the ERPP.

Part 834 specifies that the primary public dose limit include the total dose from all exposure modes and from all routine DOE activities. The primary public dose limit shall be expressed as a total effective dose equivalent (TEDE), the risk-based weighted summation of doses and dose commitments to various organs of the body.

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

In Part 834, DOE recognizes a need for a threshold level for source identification because of the inherent burden in 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 do not have to 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.

Part 834 has a requirement to report to DOE if 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.

Part 834 does not contain a collective dose limit which is the sum of the individual doses to a specified exposed population group. Collective dose is used as a reporting requirement, as a tool to help assess trends and the effectiveness of control measures, and as a tool in the ALARA process. Part 834 continues to stress the use of the ALARA decision-making process. No quantitative limits are provided but doses from DOE operations should continue to be a small fraction of the 100 mrem/yr limit.

The overall implication of Part 834 for monitoring is that DLs must be based on a dose limit 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 part 834.

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. Under the NPDES permit monitoring results at outfalls are compared to 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 into the five sections that follow.

- 2.0 Y-12 Plant Monitoring Program
- 3.0 Detection Limit Monitoring Goals



4.0	Monitoring Methodologies to Meet EPA and Tennessee Requirements
5.0	Data Management and Radiological Analysis
6.0	Quality Assurance
Appendix A	Overview Radiological Analysis of the Y-12 Plant Surface Water Radiological Monitoring Program

## 2.0 Y-12 PLANT MONITORING PROGRAM

A radiological monitoring program is in place to address compliance with DOE regulations, the NPDES Permit and the ALARA philosophy. Monitoring will be 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 these 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 which reflect individual facilities as well as at points which reflect the combined contributions of all facilities. Part 834 requirements that dose estimates consider contributions from all facilities are thus reflected in the monitoring plan for Y-12 Plant surface water. Determination of 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 incorporate other potential routes (i.e., airborne releases and food chains). Thus a complete determination of TEDE under Part 834 can not be made based on this plan alone. The other routes from the Y-12 Plant and all routes from other DOE facilities on the ORR (e.g., ORNL, K-25) must be considered in order to satisfy the requirements of Part 834. Releases from all sites are provided in the ASERs. Determination of TEDE requires the input of these releases and measurements off-site to dose assessment models. This monitoring plan for the Y-12 Plant provides adequate monitoring goals for 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 annually 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.

The state of Tennessee requirements are specified in the permit including sampling location, frequency, sample type and parameters. To meet additional state desires, Appendix A has been prepared to document 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 Steam Condensate

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. Some individual wastewater sources are analyzed for the radiological parameters discussed in Section 2.5 prior to treatment; this practice allows comparison of concentration levels before and after treatment and will aid in conducting an ALARA program for process wastes.

## 2.2 OTHER POINT AND AREA SOURCE DISCHARGES

The discharges to be monitored include Outfalls:

- 142 Isotope Separation Process at 9204-3
- S17 Kerr Hollow Quarry
- S19 Rogers Quarry

These outfalls could contribute to movement of radionuclides offsite in water that is not treated at one of the treatment facilities. The Isotope Separation Process primarily separated stable isotopes using calutrons. However, one calutron was reserved for processing small amounts of uranium isotopes ( $^{233}\text{U}$ ,  $^{235}\text{U}$ ,  $^{238}\text{U}$ ) and small amounts of some actinides (curium isotopes and  $^{242}\text{Pu}$ ).

### 2.3 INSTREAM MONITORING POINTS

The instream locations to be monitored include.

km 11.97	Kilometer 11.97 on Bear Creek
304	Bear Creek kilometer 4.55 at Highway 95
9422-1	Station 17 on East Fork Poplar Creek
9422-3	Station 8 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. Station 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 as well as provide data for mass balance evaluation of radioactive inputs. Water quality monitoring has been performed in upper Bear Creek since the early 1980's to monitor the influence of the S-3 Pond site on Bear Creek. Monitoring at Bear Creek kilometer 11.97 has been on-going since 1989 and is being continued to monitor trends.

### 2.4 SAMPLING

Previous data indicate that 7-day composites must be collected to accumulate detectable quantities of radionuclides of concern. Detection limits or minimum detectable activities for radionuclides (MDAs) 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 Energy Systems Environmental Surveillance Procedures Quality Control Program. These procedures have been approved for use by the State and EPA. Quality control checks are built into the sampling and analytical activities as discussed in Section 6. EPA has identified the analytical methods as technically and economically available for radionuclide monitoring. The NPDES Permit under Part I B 2 requires these specific 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 accuracy at 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 <sup>a</sup>	Isotopic analysis <sup>b</sup> Gamma scan <sup>c</sup>
502	West End Treatment Facility	1/week	Composite	Isotopic analysis <sup>b</sup> Gamma scan <sup>c</sup>
503	Steam Plant Wastewater Treatment Facility	1/week	Composite	Isotopic analysis <sup>b</sup> Gamma scan <sup>c</sup>
512	Groundwater Treatment Facility	1/week	Composite	Isotopic analysis <sup>b</sup> Gamma scan <sup>c</sup>
520	Steam Condensate	1/week	Grab	Isotopic analysis <sup>b</sup> Gamma scan <sup>c</sup>

<sup>a</sup>Composite will be taken during batch discharge. If facility discharges continuously, a 24-hour composite will be taken.

<sup>b</sup>Specific isotopes for NPDES and Part 834 monitoring are listed in Section 2.5.

<sup>c</sup>See text.

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
142	Isotope Separation Process	1/month	24-hour Composite	Isotopic analysis <sup>a</sup> Gamma scan <sup>b</sup>
S17	Kerr Hollow Quarry	1/month	24-hour Composite	Isotopic analysis <sup>a</sup> Gamma scan <sup>b</sup>
S19	Rogers Quarry	1/month	24-hour Composite	Isotopic analysis <sup>a</sup> Gamma scan <sup>b</sup>

<sup>a</sup>Specific isotopes for NPDES and Part 834 monitoring are listed in Section 2.5.

<sup>b</sup>See text.

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 <sup>a</sup> Gamma scan <sup>b</sup>
17 9422-1	East Fork Poplar Creek	1/week	7-day Composite	Isotopic analysis <sup>a</sup> Gamma scan <sup>b</sup>
8 9422-3	East Fork Poplar Creek, Plant Site	1/week	7-day Composite	Isotopic analysis <sup>a</sup> Gamma scan <sup>b</sup>
200	North/South Pipes	1/week	24-hour Composite	Isotopic analysis <sup>a</sup> Gamma scan <sup>b</sup>
km 11.97	Bear Creek	1/week	grab	Isotopic analysis <sup>a</sup> Gamma scan <sup>b</sup>

<sup>a</sup>Specific isotopes for NPDES and Part 834 monitoring are listed in Section 2.5.

<sup>b</sup>See text.

The NPDES permit (Part I) requires measurement frequencies of 1/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 4 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 1/week or 1/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 current version of the RMP therefore includes gamma scans. The results will be evaluated and reported to the Division of Water Pollution Control by August 1, 1996. If the results confirm previous operational history and monitoring results, gamma scanning may be eliminated from the RMP.



## 2.5 RADIOLOGICAL PARAMETERS MONITORED

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

$^{238}\text{U}$

$^{235}\text{U}$

$^{234}\text{U}$

and total U and weight percent  $^{235}\text{U}$

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 1994. The isotopes  $^{234}\text{Th}$  and  $^{234}\text{Pa}^m$  will be in secular equilibrium with  $^{238}\text{U}$  unless there are processes in operation which enrich them relative to  $^{238}\text{U}$  with direct release to surface water.

Fission and activation products to be monitored include:

$^{90}\text{Sr}$

$^3\text{H}$

$^{99}\text{Tc}$

$^{137}\text{Cs}$

These parameters reflect a relatively minor activity at the Y-12 Plant - processing of recycled uranium from reactor fuel elements in the early 1960s. Conservative analysis of expected relative isotopic concentrations assuming direct processing of fuel elements indicate  $^{90}\text{Sr}$  concentrations potentially higher than other fission products. The isotope  $^3\text{H}$  is not expected to be high in fuel elements - it is produced primarily as an activation product in reactor coolants. But  $^3\text{H}$  is highly mobile and is detected in groundwater samples associated with the S-3 Pond Site. The isotope  $^{99}\text{Tc}$  is also expected to be at relatively low concentrations as a fission product in reactor fuel elements. However,  $^{99}\text{Tc}$  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  $^{99}\text{Tc}$  and  $^3\text{H}$  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 has 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 compared to  $^{90}\text{Sr}$ . Gamma spectrometry is included to detect any significant concentrations of gamma emitters that may have been missed in this analysis.

Transuranium isotopes to be monitored include:

$^{241}\text{Am}$  -  
 $^{237}\text{Np}$  ( $^{233}\text{Pa}$ )  
 $^{238}\text{Pu}$   
 $^{239}\text{Pu}$

These isotopes are also related to recycle uranium processing. Concentrations were estimated to be low in fuel elements compared to  $^{90}\text{Sr}$  but toxicities are much higher. They are expected to be less mobile than  $^{90}\text{Sr}$ . Concentrations have been low in surface water but they are monitored because of their long half lives and long term risk potential. Some  $^{237}\text{Np}$  groundwater results have been suspect because expected equilibrium with  $^{233}\text{Pa}$  was not observed. Analysis for  $^{233}\text{Pa}$  should thus continue until the suspect results are fully explained or shown to be erroneous.

Isotopes associated with thorium processing and other natural radionuclides include:

$^{232}\text{Th}$   
 $^{230}\text{Th}$   
 $^{228}\text{Th}$   
 $^{226}\text{Ra}$   
 $^{228}\text{Ra}$

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). Anomalous results for  $^{228}\text{Ra}$  were corrected by changing the sample analysis procedure (see Section 5.2).

## 2.6 STORM WATER MONITORING

The NPDES permit also requires (Part III H) that the RMP be expanded to include storm water monitoring. A draft 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 characterize 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 every time 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 draft 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 will be conducted. Data will be reported, as acquired, to the TDEC as an attachment to the DMR.

The initial data obtained in the Storm Water Monitoring Plan will be included in the assessment and reevaluation of the RMP by August 1, 1996 (see Section 2.4). This assessment and re-evaluation may provide a basis for modification and/or refinement of the storm water monitoring component of the RMP.

### 3.0 DETECTION LIMITS AND MONITORING GOALS

Detection limits or minimum detectable activities (MDAs) required vary according to the purpose of the activity. Detection limits required to meet monitoring goals based on NPDES permit requirements, Part 834 regulations, and preelimination remediation goals under development by the on going environmental restoration projects are discussed in this section. More detail may be found in Section 3 of Appendix A.

#### 3.1 CONCENTRATION LIMITS FOR MONITORING GOALS

Under current conditions at the Y-12 Plant, surface water is not a source of public water supply before it reaches the Clinch River, where effluents are greatly diluted before any use of the water for a public water supply. 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, K-25 and ORNL.

Under the current DOE guidance, radioisotopes should be monitored at concentrations about 0.2 DCG (see Section 3.2 and Appendix A for further discussion). This result is based on water concentration in East Fork Poplar Creek and Bear Creek that meet 4 mrem/yr drinking water criteria in the Clinch River. The 4 mrem/yr dose is not taken as a regulatory limit for all isotopes. However, its use will ensure that drinking water limits are not exceeded for any isotope.

Reference concentrations for Y-12 Plant radionuclides based on DCFs from the latest EPA guidance (Federal Guidance Report Number 11, Eckerman et al. 1988) and the 4 mrem/yr limit are given in Table 4. The reference concentrations may also be obtained, to good approximation, by dividing the DOE DCGs (DOE Order 5400.5), which correspond to a 100 mrem/yr dose, by 25. There are minor differences because the DCFs have been updated in the EPA guidance. The DOE DCGs from DOE Order 5400.5 are given in Table 4. 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.

Table 4. Dose conversion factors and limiting concentrations for Y-12 radiological contaminants

Nuclide	Ingestion Dose Conversion Factor (rem/ $\mu$ ci)	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

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\* 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.

Thus, the sum rule for concentrations is

$$\sum C_i/CL_i < 1$$

where  $C_i$  is the concentration of radioisotope  $i$  in a particular environmental pathway and  $CL_i$  is the concentration limit for radioisotope  $i$  in that pathway.

The sum rule is stated in 10 CFR 834.211 (c) (1) as follows:

"For purposes of this section, if more than one type of radionuclide A, B, ....N are present in concentrations  $C_A$ ,  $C_B$ , .... $C_N$ , and if applicable derived concentration guide values are  $DCG_A$ ,  $DCG_B$ , .... $DCG_N$  respectively, then the concentration shall be limited so that the following relationship is satisfied.

$$(C_A/DCG_A) + (C_B/DCG_B) + \dots + (C_N/DCG_N) \leq 1"$$

The sum rule is stated in the primary public dose limit under Part 834 as: "Radiation exposures to individuals in the general public from all radiation sources and exposure pathways combined from routine DOE activities shall not exceed 100 mrem TEDE in a year."

Typical MDAs reported by the Energy Systems Analytical Services Organization (ASO) are below drinking water criteria for each isotope (see Appendix A, Table 5). Therefore, they are adequate for regulatory purposes. However, concentrations below MDAs are often encountered and MDAs will have to be reduced to maintain the desired 95 percent confidence level.

Total U concentrations based on isotopic composition are often significantly higher than total U concentrations based on mass spectrometry. The differences are probably due to interferences among isotopes when alpha spectrometry is used. Measurement of total U based on mass spectrometry should continue. However, the monitoring goal for  $^{235}\text{U}$  at 1 ppb total U will have to be about 0.01 pCi/L (0.005 ppb) in order to obtain weight percent  $^{235}\text{U}$ . The activity concentration of  $^{234}\text{U}$  could be measured instead of  $^{235}\text{U}$ , in which case the MDA would have to be about 0.1 pCi/L at 1 ppb total U (see Appendix A, Table 1).

Such low monitoring goals would be adequate for practical quantification far below that necessary for regulatory purposes. Such low monitoring goals also provide a basis for confidently excluding radioisotopes with measured concentrations below monitoring goals from further monitoring. However, current typical MDAs are not adequate to meet these monitoring goals without costly improvements.

The detection limits or MDAs necessary to meet monitoring goals based on Part 834 and

NPDES requirements along with current practical quantitation levels are given in Table 5.

The current regulatory column 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 criteria (1/25 DCGs). These goals are adequate for regulatory purposes but not adequate to monitor current levels (See Section 3.2). They are about 3 times lower than the 0.2 DCG level discussed above and in Section 3, Appendix A. They ensure that concentrations at the MDAs can be determined with 95 percent confidence.

The preliminary 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 quantitative 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 ALARA. They are not necessary to demonstrate regulatory compliance but would reduce the need for special studies requiring MDAs lower than the regulatory requirements.

### 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 to their DCGs in the ASERs. At both locations, all isotopes except for  $^{234}\text{U}$  and  $^{238}\text{U}$  were at concentrations less than 1 percent of their DCGs.  $^{234}\text{U}$  and  $^{238}\text{U}$  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 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. Other facilities in the ORR contribute more to the drinking water route than does the Y-12 Plant but are still well below drinking water criteria.

Table 5. Surface water monitoring goals for Y-12 Plant radionuclides

Parameters	Monitoring Goals (pCi/L)	
	Current NPDES <sup>1</sup>	Practical Quantitation Levels <sup>2</sup>
U-238	30	0.5
U-235	37.5	0.5
U-234	37.5	0.5
Th-228	25	0.5
Th-230	20	0.5
Th-232	3	0.5
Ra-226	6	1.0
Ra-228	0.6	3.0
Pu-238	2	1.0
Pu-239	2	1.0
Np-237	1.9	0.8
Am-241	0.2	0.5
Tc-99	6.25E+03	8.0
Sr-90	62.5	15
H-3	1.25E+05	400
Cs-137	1.70E+02	6.0

<sup>1</sup> Sum of fractions = 1 where limit is DCG for each isotope

<sup>2</sup> Typical detection limits currently reported by the Analytical Services Organization (See Table 5, Appendix A).



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. Taking into consideration releases from other sites and other pathways under Part 834, 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 (0.06) 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 were about 6.2 percent and about 8 percent for all isotopes combined. The concentration of each isotope was compared to its DCG.

Based on these results reporting to DOE under Part 834 will not be required and the effort required to prepare an ERPP should be low in accord with the extremely low risk associated with the drinking water pathway.

Radionuclide releases to Y-12 Plant surface water also make a relatively small contribution to terrestrial and aquatic food chains.

Thus, this monitoring plan, required by the NPDES permit, may serve as input to the ERPP required under Part 834 which must incorporate all pathways and all facilities.

In order to meet the proposed requirements of Part 834, the radiological assessments in the ASERs will need to be oriented towards exposure to members of the public by all pathways as opposed to the current orientation of maximal exposure locations which differ for each pathway.

#### 4.0 MONITORING METHODOLOGIES TO MEET EPA AND TENNESSEE REQUIREMENTS

In this section, the specific instrumental sampling and 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 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 overall DOE policy with respect to DOE versus EPA or state requirements is stated in Section 5.2.4 of the DOE Regulatory Guide as follows.

"DOE Operations Office and contractor staff should\* ensure that groundwater monitoring plans are consistent with state and required EPA groundwater monitoring requirements under RCRA and CERCLA to avoid unnecessary duplication. DOE Operations Office and Contractor staff should\* consult with state and required EPA offices, as needed, to ensure that the requirements are incorporated into the Radiological Monitoring Plan."

This policy applies to the NPDES requirements as well. 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 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.

The methods listed in Table 6 may be applied according to the procedures given in the list of references for Table 6 for surface water at Y-12. The monitoring system has the capability to monitor to levels below the requirements of Part 834, demonstrate improvement, 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	Y-12 Procedure	Reference
Gross alpha and beta	Evaporation/Gas Flow Proportional Counting	Y/P65-7162	EPA 900.0 <sup>1</sup>
Total Alpha Radium	Barium Sulfate Coprecipitation Gas Flow Proportion Counting	Y/P65-7163	EPA 903.0 <sup>1</sup>
Radium-228	Barium and Lead Sulfate Coprecipitation and <sup>228</sup> Ac Ingrowth Gas Flow Proportional Counting	Y/P65-7163	EPA 904.0 <sup>1</sup>
Americium-241	Neodymium fluoride coprecipitation and separation by anion exchange/Alpha Spectrometry	Y/P65-7157	2, 3, 4
Neptunium-237	Neodymium fluoride coprecipitation and separation by anion exchange/Alpha Spectrometry	Y/P65-7158	3, 4, 5
Plutonium-238,239/240	Neodymium fluoride coprecipitation and separation by anion exchange/Alpha Spectrometry	Y/P65-7159	3, 4, 6
Thorium-228,232,230	Neodymium fluoride coprecipitation and separation by anion exchange/Alpha Spectrometry	Y/P65-7160	3, 4, 7
Uranium Isotopic	Neodymium fluoride coprecipitation and separation by anion exchange/Alpha Spectrometry	Y/P65-7161	3, 4, 8
Total Uranium and % U-235	Solvent Extraction/Thermal Ionization Mass Spectrometry	Y/P65-7165	8
Cesium-137	Gamma Spectrometry	Y/P65-7171	EPA 901.1 <sup>1</sup>
Strontium 89/90	Separation by chromatographic resin, <sup>90</sup> Y ingrowth, Gas Flow Proportional Counting	Y/P65-7196	EPA 905.0 <sup>1</sup>
Technetium-99	Separation by Iron Coprecipitation Liquid Scintillation Counting	Y/P65-7154	9
Tritium	Distillation/Liquid Scintillation Counting	Y/P65-7150	EPA 906.0 <sup>1</sup>

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  $^{241}\text{Am}$ ,  $^{244}\text{Cm}$ ,  $^{237}\text{Np}$ ,  $^{238}\text{Pu}$ ,  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$ ,  $^{228}\text{Th}$ ,  $^{230}\text{Th}$ ,  $^{232}\text{Th}$ ,  $^{234}\text{U}$ ,  $^{235}\text{U}$ ,  $^{236}\text{U}$ , and  $^{238}\text{U}$ . 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  $^{234}\text{Pa}$ ,  $^{137}\text{Cs}$ ,  $^{60}\text{Co}$ ,  $^{106}\text{Ru}$ . 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. Gamma emitters have been below MDAs at the Y-12 Plant and thus do not make a significant contribution to TEDE under Part 834.

Concentrations of  $^{137}\text{Cs}$  have been below a MDA of about 10 pCi/L compared to a 4 mrem/yr concentration of 110 pCi/L.  $^{106}\text{Ru}$  concentrations have been below a MDA of about 40 pCi/L compared to a 4 mrem/yr concentration of 200 pCi/L.

The only individual beta emitters currently monitored at the Y-12 Plant are  $^{90}\text{Sr}$ ,  $^3\text{H}$  and  $^{99}\text{Tc}$ . The isotopes  $^3\text{H}$  and  $^{99}\text{Tc}$  are determined by liquid scintillation counting. Sample volumes are 0.05L for  $^3\text{H}$  and 0.4L for  $^{99}\text{Tc}$ . A counting time of 1 hour results in DLs of 400 pCi/L for  $^3\text{H}$  and 8 pCi/L for  $^{99}\text{Tc}$  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 specifically regulated in drinking water supplies under the Safe Drinking Water Act and are detected in groundwater.

Similarly,  $^{90}\text{Sr}$  is monitored because it is specifically 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 to the EPA MCL of 8 pCi/L (see Table 4).

#### 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 3.0 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,  $R(\text{total})$ , 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  $R(\text{total})$  can lead to negative numbers with large error especially if  $R_B$  does not include the same matrix effects as  $R(\text{total})$ .

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 USEPA 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

$$V_i = 100 \sigma_i / R_i$$

is the coefficient of variation (percent error) of  $R_i$  and  $\sigma_i$  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, while  $R(\text{total})$  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.

$$R_{S+B} = R_S + R_E + R_B$$

The net sample count is therefore not simply  $R(\text{total}) - R_B$  but  $R(\text{total}) - R_E - R_B$ . 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 Y-12 Plant ASOs. 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 the EPA. Concentrations of alpha-, gamma-, and beta-emitting radionuclides in Y-12 Plant surface waters are generally low compared to 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.

## 5.0 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 Environmental Laboratory Information Management System (ELIMS) on the same day it is obtained. ELIMS will be electronically accessed on a routine basis by EMMIS. EMMIS will capture pertinent data in ELIMS 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 to regulatory and best management or ALARA goals and produce required reports. The state of Tennessee requires that initial comparisons be made with DCGs. The surface water subsystem of EMMIS is currently being programmed and implemented. Radiological data, in addition to other types of data, will also be transferred into EMMIS where it will be readily retrievable for use in reports or for special statistical analysis.

### 5.2 RADIOLOGICAL ANALYSIS METHODOLOGY WITH EXAMPLES

The Y-12 Plant surface water radiological data bases have been analyzed for two reasons: (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 may be found in 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  $^{235}\text{U}$ . Radioisotopes to be considered from these activities are  $^{238}\text{U}$ ,  $^{235}\text{U}$ , and  $^{234}\text{U}$ . In addition, some 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 cycle reactor fuel elements were not processed.

On the basis of these general activities, surface water monitoring at the Y-12 Plant has included alpha, beta, and gamma screening; the uranium isotopes  $^{234}\text{U}$ ,  $^{235}\text{U}$ , and  $^{238}\text{U}$ ; weight percent enrichment of  $^{235}\text{U}$ ; the fission products  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$ ,  $^3\text{H}$ ,  $^{90}\text{Sr}$ , and  $^{99}\text{Tc}$ ; the transuranics  $^{241}\text{Am}$ ,  $^{237}\text{Np}$ ,  $^{238}\text{Pu}$ , and  $^{239}\text{Pu}$ ; and biological tracers such as  $^{14}\text{C}$ ,  $^{35}\text{S}$ ,  $^{32}\text{P}$ ,  $^{129}\text{I}$ , and  $^{125}\text{I}$ . 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  $^{234}\text{U}$ ,  $^{235}\text{U}$ ,  $^{238}\text{U}$ ,  $^{237}\text{Np}$ ,  $^{241}\text{Am}$ ,  $^{228}\text{Ra}$ ,  $^{226}\text{Ra}$ ,  $^{228}\text{Th}$ ,  $^{230}\text{Th}$ ,  $^{232}\text{Th}$ ,  $^{234}\text{Th}$ ,  $^{99}\text{Tc}$ ,  $^{90}\text{Sr}$ , and  $^3\text{H}$  in addition to alpha, beta and gamma screening measurements. Total uranium and weight percent  $^{235}\text{U}$  are also monitored. The 1991, 1993 and 1994 results for selected outfalls 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.

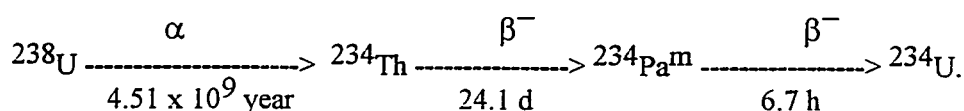
The monitoring history of the Y-12 Plant, DOE, EPA, and Tennessee requirements, and the methodology outlined herein provide a rationale for the radiological parameters to be included in the revised Radiological Monitoring Plan.

In addition to continued comparison of the data to 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, the time history is known, and 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 suspect until and unless 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 are described in the following two examples.

- (1) Concentrations (in activity per unit volume) of  $^{234}\text{Th}$  and  $^{234}\text{Pa}^{\text{m}}$  should be approximately the same as concentrations of  $^{238}\text{U}$  because the relative radiological half lives would produce secular equilibrium in a matter of months. Unless the uranium entering the processes at the Y-12 Plant is free of thorium and protactinium and moves through the processes in a matter of days to weeks, then secular equilibrium can be expected according to the partial decay chain for  $^{238}\text{U}$ .



Monitoring for  $^{238}\text{U}$  will allow conservative estimation of  $^{234}\text{Th}$  and  $^{234}\text{Pa}^m$  by assuming equilibrium. Their concentrations cannot exceed that of  $^{238}\text{U}$  unless additional sources, concentration, or separation processes can be identified that are currently being released to surface water. In any case  $^{234}\text{Th}$  and  $^{234}\text{Pa}^m$  unsupported by  $^{238}\text{U}$  would decay in a matter of months. Analyses of data for  $^{234}\text{U}$  and  $^{238}\text{U}$  in surface water at the Y-12 Plant indicate that  $^{238}\text{U}/^{234}\text{U}$  ratios are consistent and are those expected on the basis of percent enrichment in  $^{235}\text{U}$ . Note that monitoring of  $^{234}\text{U}$  is necessary to support such analysis. Monitoring of  $^{235}\text{U}$  and/or percent enrichment in  $^{235}\text{U}$  is also necessary. Given the consistency and reliability of the uranium measurements, lack of equilibrium of measured  $^{234}\text{Th}$  with  $^{238}\text{U}$  flags the 1991  $^{234}\text{Th}$  results as suspect. The 1993 and 1994 results for  $^{234}\text{Th}$  are based on equilibrium with  $^{238}\text{U}$ .

- (2) Similarly to the first example, the concentrations of  $^{232}\text{Th}$ ,  $^{228}\text{Ra}$ , and  $^{228}\text{Th}$  should be approximately the same for the time period since thorium processing occurred at the Y-12 Plant. According to analysis of the 1991 Y-12 Plant monitoring data, concentrations in surface water do not consistently show equilibrium singly or in combination. The  $^{228}\text{Ra}$  values are particularly suspect because they are often much higher than  $^{232}\text{Th}$  even though no other source for  $^{228}\text{Ra}$  has been identified. Ratios of  $^{232}\text{Th}$  to  $^{228}\text{Th}$  are also highly variable and almost always less than 1 (one). The ratios are often as low as 0.01. These low ratios are, at least in part, accounted for by the fact that  $^{232}\text{U}$  is used as a tracer in the analytical procedures for uranium. Equilibrium between  $^{228}\text{Th}$  and  $^{232}\text{U}$  would be established in about 5 years so that the  $^{228}\text{Th}$  "growing in" from the  $^{232}\text{U}$  tracer could make a major contribution to the total  $^{228}\text{Th}$  count. Since the  $^{232}\text{U}$  decays directly to  $^{228}\text{Th}$ , the  $^{228}\text{Ra}$  is unaffected. Thus the high  $^{228}\text{Ra}/^{232}\text{Th}$  ratios for 1991 remain outside expectations.

If the elevated 1991  $^{228}\text{Ra}$  numbers are reliable, then  $^{228}\text{Ra}$  concentrations are very erratic both in surface water and groundwater.  $^{232}\text{Th}$  contamination with a periodic flushing of  $^{228}\text{Ra}$ , perhaps by a storm event, might explain the erratic nature of the  $^{228}\text{Ra}$  concentrations without the presence of equally elevated  $^{232}\text{Th}$  and  $^{228}\text{Th}$ . Differential solubility of Ra versus Th (Th being much less soluble) and then turnover of  $^{228}\text{Ra}$  via surface water or groundwater turnover before ingrowth of  $^{228}\text{Th}$  could account for the high  $^{228}\text{Ra}$  values relative to both  $^{232}\text{Th}$  and  $^{228}\text{Th}$  in some Y-12 Plant samples.

The  $^{228}\text{Ra}$  analytical technique used in 1991 was not sensitive enough and probably not accurate enough to reach any quantitative conclusions at this time. Laboratory analysis for  $^{228}\text{Ra}$  that has yielded variable results involves allowing  $^{228}\text{Ac}$  to "grow in" and then using gamma spectroscopy to quantify  $^{228}\text{Ac}$  and indirectly  $^{228}\text{Ra}$  since it should be in equilibrium with  $^{228}\text{Ac}$ . A standard reference source containing multiple gamma emitters is used to calibrate the gamma spectrometry system. Actual field samples potentially contain numerous other gamma emitters that may

contribute under the  $^{228}\text{Ac}$  peaks to a different degree than the reference source. One example is  $^{233}\text{Pa}$ . It emits a complex gamma spectrum including 41% occurrence of a 0.31 MeV gamma.

The energies of the gammas utilized to quantify the  $^{228}\text{Ac}$  are 0.34, 0.908, and 0.96 MeV. Other isotopes that could contribute to the  $^{228}\text{Ac}$  gamma peak areas include  $^{237}\text{U}$  (1.4%, 0.332 MeV),  $^{154}\text{Eu}$  (12%, 0.876 MeV; 31% doublet, 1.00 MeV among others),  $^{94}\text{Nb}$  (100%, 0.871 MeV),  $^{54}\text{Mn}$  (100%, 0.835 MeV), and  $^{26}\text{Al}$  (4%, 1.12 MeV). However, these isotopes are expected to be very low in Y-12 Plant surface water.

The short-lived radon daughters ( $^{218}\text{Po}$ ,  $^{214}\text{Bi}$ ,  $^{214}\text{Po}$ ,  $^{214}\text{Pb}$ ) emit a complex gamma spectrum that could interfere with the analysis of  $^{228}\text{Ac}$ . Calibration of a gamma spectrometry system to accurately analyze for low levels of  $^{228}\text{Ra}$  in such a potentially complex background is very difficult. The alternate method specified by the EPA (see Section 4), which involves initial precipitation of Ra, should be much more accurate. Sensitive and accurate analysis for low levels of specific isotopes in mixtures will generally require initial separation by radiochemical techniques. The 1993 and 1994 results using the method listed in Table 6 are substantially in agreement with expectations taking into account use of  $^{232}\text{U}$  as a tracer and differential solubility of Ra versus Th. The 1993 and 1994  $^{228}\text{Ra}$  values are very low compared to the 1991 results.

## 6.0 QUALITY ASSURANCE

In May 1995, DOE issued an update of 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 Facilities" (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 Oversight 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

#### 1.0 INTRODUCTION

The state of Tennessee Division of Water Pollution Control has communicated their needs and goals for radiological monitoring plans (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):

- Monitoring of outfalls which have potential to discharge radioactivity to waters of the state.
- Mechanism for information about radiological parameters to be collected and made available to the public.
- 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 required detection limits 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 general issues pertinent to documentation of the basis for the surface water RMP for the Y-12 Plant. Updates of the RMP will address specific issues associated with each plan.

A wide array of radioisotopes have been monitored and alpha, beta and gamma screening measurements have been conducted. The monitoring history of the Y-12 Plant and DOE, EPA and Tennessee requirements have resulted in procedure changes over the years and to changes in radioisotopes and sites monitored that are not based on readily available documented analysis. 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 supporting 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 the Y-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 then 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 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 needs to 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. In a longer time frame, the analysis needs to be supported with more in-depth analysis and detail.

## 2.0 IDENTIFICATION OF RADIOISOTOPES

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 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. Storage below ground level or in protected surface facilities generally precludes a runoff contribution to surface water.

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  $^{235}\text{U}$ . Some recycle uranium from reactor fuel elements containing fission products and transuranics was processed from the early 1960s to 1988. Some 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 continues to be collected and is being evaluated for risk assessment purposes by the Y-12 Environmental Restoration Program. Data is also reported in the ASERs, in NPDES reports and in Groundwater Quality Assessment Reports.

## 2.1 Uranium Processing

Since  $^{230}\text{Th}$  and  $^{226}\text{Ra}$  in the  $^{238}\text{U}$  decay series, along with corresponding radioisotopes in the  $^{235}\text{U}$  series were removed in the milling component of the uranium fuel cycle, the  $^{238}\text{U}$ ,  $^{235}\text{U}$  and  $^{234}\text{U}$  isotopes and relatively short lived daughters are the primary radioisotopes of monitoring interest. The  $^{232}\text{Th}$  chain was also removed during milling. The  $^{238}\text{U}$  daughters products,  $^{234}\text{Th}$  and  $^{234}\text{Pa}^{\text{m}}$ , occur up to concentrations equal to  $^{238}\text{U}$  because secular equilibrium will be established in months. The concentrations of  $^{234}\text{Th}$  and  $^{234}\text{Pa}^{\text{m}}$  will not exceed  $^{238}\text{U}$  concentrations unless there is some differential concentration mechanism for  $^{234}\text{Th}$  and  $^{234}\text{Pa}^{\text{m}}$  or dilution mechanism for  $^{238}\text{U}$ . Plant processes or liquid waste treatment systems could remove uranium isotopes and leave higher concentrations of  $^{234}\text{Th}$  and  $^{234}\text{Pa}^{\text{m}}$  in liquid wastes. However, since these radioisotopes decay with a half life of about 24 days, higher concentrations of  $^{234}\text{Th}$  and  $^{234}\text{Pa}^{\text{m}}$  unsupported by  $^{238}\text{U}$  should have substantially decayed 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  $^{235}\text{U}$  by the gaseous diffusion process will have a predictable amount of  $^{234}\text{U}$  and  $^{238}\text{U}$ . If monitoring results are in agreement with predictions, then other processes such as discussed above for  $^{234}\text{Th}$  and  $^{234}\text{Pa}^{\text{m}}$  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  $^{235}\text{U}$ .

Uranium analysis can be reliably based on total U analysis on a mass basis combined with a separate determination of  $^{235}\text{U}$  in order to determine percent mass enrichment of  $^{235}\text{U}$ . These determinations will allow calculation of the specific activity of total U and an estimate of the isotopic composition of total U if no other significant source of  $^{234}\text{U}$  exists. When total U is less than 1 ppb, the sensitivity for determining %  $^{235}\text{U}$  will need to be improved because the specific activity for total U at 1 ppb mass

concentration can vary from 0.67 pCi/μg for natural U or less for depleted U to 67 pCi/μg for 95 percent enrichment.

In the past, total U estimates based on activity concentrations of  $^{238}\text{U}$ ,  $^{235}\text{U}$  and  $^{234}\text{U}$  have been significantly higher than total U 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  $^{234}\text{U}$ ,  $^{235}\text{U}$  and  $^{238}\text{U}$ . 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 ( $^{235}\text{U}$  less than 0.72 weight percent) so that measurement of total U and calculation of isotopic composition assuming natural U would overestimate the concentrations of  $^{235}\text{U}$  and  $^{234}\text{U}$  and underestimate the concentration of  $^{238}\text{U}$ . However, there are enough exceptions where the uranium appears to be enriched so that total U analysis alone is not sufficient to determine isotopic activities. If the calculation of total U specific activity were based on  $^{234}\text{U}$ , then the detection limit requirement could be substantially relaxed. The concentration of  $^{234}\text{U}$  for 1 ppb natural U is about 0.33 pCi/L. The concentration of  $^{234}\text{U}$  in total U at 1 ppb enriched to 95 percent in  $^{235}\text{U}$  is about 65 pCi/L. For natural uranium, the concentration of  $^{235}\text{U}$  at 1 ppb total U would be 0.014 pCi/L. For 95 percent enriched uranium the  $^{235}\text{U}$  concentration is 0.95 ppb or 1.9 pCi/L.

The isotopic compositions at various levels of enrichment in  $^{235}\text{U}$  for a total U concentration of 1 ppb are given in Table 1. For other levels of total U, each column in

Table 1. Isotopic composition of a 1 ppb total U water sample versus weight percent of  $^{235}\text{U}$ .

Weight % $^{235}\text{U}$	$^{235}\text{U}$ (ppb)	Total U (pCi/L)	$^{238}\text{U}$ (pCi/L)	$^{234}\text{U}$ (pCi/L)	$^{235}\text{U}$ (pCi/L)
0.1	0.001	0.438	0.327	0.109	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

Table 1 except the first column would be multiplied by the total U content in ppb. A  $^{234}\text{U}$  detection limit of 0.1 pCi/L is adequate to fully characterize uranium isotopes based on total U and  $^{234}\text{U}$  analysis.

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

- a) Use Table 1 to determine weight %  $^{235}\text{U}$  from  $^{234}\text{U}$  measurement.
- b) Calculate specific activity (SPA) of total U from Rich formula (Rich et al, 1988).

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

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

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

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

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

The required measurements are total U with a detection limit of 1 ppb and  $^{234}\text{U}$  with a detection limit of 0.1 pCi/L for depleted U. Expected equilibrium of  $^{234}\text{Th}$  and  $^{234}\text{Pa}^m$  with  $^{238}\text{U}$  provides for their estimation. Possible interferences with  $^{234}\text{U}$  measurements must be removed by radiochemical techniques for accurate analysis. Possible interfering isotopes are discussed further for transuranium isotopes in Section 2.2.

Early monitoring results for Y-12 Plant surface water included only total U and weight %  $^{235}\text{U}$ . These results were adequate given the assumption that  $^{234}\text{Th}$  and  $^{234}\text{Pa}^m$  are in equilibrium with  $^{238}\text{U}$ . More recent measurements have included  $^{234}\text{Th}$

and  $^{234}\text{Pa}^m$  with results sometimes indicating  $^{234}\text{Th}$  and  $^{234}\text{Pa}^m$  at levels higher than  $^{238}\text{U}$ . However, as discussed in Section 5.2 of the RMP, the elevated  $^{234}\text{Th}$  and  $^{234}\text{Pa}^m$  results relative to  $^{238}\text{U}$  are suspect unless there is a current source for  $^{234}\text{Th}$  and  $^{234}\text{Pa}^m$ . 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  $^{234}\text{U}$  concentrations are generally less than  $^{238}\text{U}$  concentrations. Since the half life of  $^{234}\text{U}$  is very long compared to its parents except for  $^{238}\text{U}$ , the  $^{234}\text{U}$  present is due to decay from  $^{238}\text{U}$  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. Surface water concentrations resulting from surface runoff are relatively low. A storm water monitoring plan is in place to determine the contribution of storm water runoff to radioisotopes in surface water. A summary of the plan is given in Section 2.6 of the RMP.

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 identification of isotopes which could be monitored. Knowledge that the reactor was fueled by  $^{235}\text{U}$  is enough to roughly approximate the relative amounts of fission products produced per gram of  $^{235}\text{U}$  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  $^{235}\text{U}$  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 3 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  $^{235}\text{U}$  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. To the amounts corresponding to the yields listed in Table 1 has been added the contributions from the decay of other isotopes to the isotopes listed. For example to the amount calculated from the fission yield for  $^{137}\text{La}$  is added

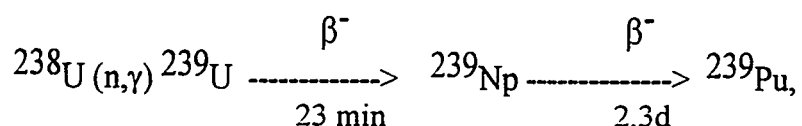
Table 2. Fission products contributing more than 1 mCi per gram of  $^{235}\text{U}$  consumed 30 years after removal from a reactor

Isotope	Yield (%) Approx.	Half Life (years)	Activity per gram $^{235}\text{U}$ (Ci)	Emission Energy (MeV)		
				$\alpha$	$\beta$	$\gamma$
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 (100%)				0.935	0
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 Ag-108 (79%)				0.61	0.018
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
Sn-121	Secular equilibrium with Sn-121m (77%)				0.114	0
Cs-134	6	2.062	1.60E-03	0	0.164	1.555
Cs-137	6	30	2.57	0	0.187	0
Ba-137m	Secular equilibrium with Cs-137 (94.6%)				0.0655	0.597
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



$^{137}\text{La}$  from the decay of  $^{137}\text{Ce}$  and  $^{137}\text{Pr}$  which are produced at about the same fission yield as  $^{137}\text{La}$ .

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



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

The transuranic 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 of 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.  $^{90}\text{Sr}$  among the fission products) then the overall result could be extremely conservative. A significant fraction of the historic data base 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.

Currently, highly specific ion exchange resins are available for specific isotopes and advances are continuous. The samples are spiked with a known amount of the specific isotope analyzed so that recovery and system response to a known amount can be determined.

The Analytical Services Organization (ASO) uses EPA-approved procedures or better for most of the analyses. However, they are also using other procedures for more quantitative results when available including the ion exchange resins.

In the past, consistent procedures have not always been utilized among the three major sites on the ORR. The ASO is currently consolidating the needs of all three sites at a central location. This centralization is intended to help to ensure consistent up-to-date procedures for all ORR sites.

Identification of isotopes which 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  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$ , and  $^3\text{H}$  tend to disperse more than other fission products in water. They are produced in higher quantities than the other fission products.  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  have relatively high dose conversion factors and relatively long half lives.

Based on this general knowledge,  $^{90}\text{Sr}$  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  $^{90}\text{Sr}$  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  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  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  $^{90}\text{Sr}$ . It is also assumed that all fission and activation products have the same relative concentrations in wastes as in fuel elements. 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.

Table 3. Relative hazards of fission radioisotopes in a representative reactor fuel element

Isotopes	Ci/Ci $^{90}\text{Sr}$	Ingestion DCFi/DCF $^{90}\text{Sr}$	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

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.  $^{137}\text{Cs}$  or Europium isotopes)  $^{90}\text{Sr}$  and  $^3\text{H}$  monitoring on quarterly composites for surface water and add  $^{226}\text{Ra}$  on annual composites for groundwater.

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

Past monitoring has included  $^{60}\text{Co}$ ,  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$ ,  $^3\text{H}$  and  $^{99}\text{Tc}$ . The inclusion of  $^3\text{H}$  and  $^{99}\text{Tc}$  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. Monitoring for  $^{60}\text{Co}$  has been discontinued because, as expected, concentrations relative to  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  have been very low.

Selection of radioisotopes to be monitored in surface water depends on Y-12 Plant operational history including waste management practices and review of groundwater, surface water and storm water data.

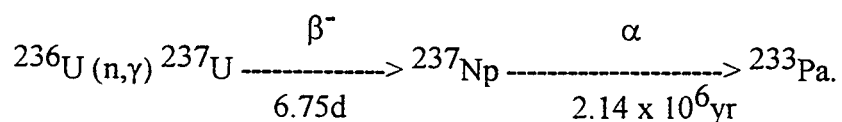
Review of groundwater and surface water data for the Y-12 Plant indicates that the concentrations of fission products are generally low relative to the uranium isotopes.

The only isotopes that exceed 4 mrem/yr drinking water criteria in the most contaminated on-site wells near the S-3 Site are  $^{99}\text{Tc}$  and  $^{90}\text{Sr}$ . Concentrations of  $^{90}\text{Sr}$  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  $^{90}\text{Sr}$  concentrations are about 0.1 - 0.2 percent of the DCG (about 2.5 - 5 percent of drinking water criteria).

The indication that  $^{90}\text{Sr}$  may be the key fission product radioisotope based on qualitative analysis is supported by the monitoring data. 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  $^{90}\text{Sr}$  and have very long half lives. Thus, they can represent a long term hazard much greater than  $^{90}\text{Sr}$  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. Since production of these isotopes vary with fuel enrichment, it has been important to monitor for them.

Other isotopes may be important because of their amount and toxicity but also because they may interfere with analyses for other isotopes. The isotope  $^{237}\text{Np}$ , the daughter product of  $^{241}\text{Am}$  can also be produced via the reaction



To a lesser extent  $^{237}\text{U}$  will be produced by  $^{238}\text{U} (n, 2n)$  and  $^{235}\text{U} (2n,\gamma)$  reactions. Concentrations of  $^{237}\text{Np}$  could be higher than  $^{241}\text{Am}$  in spite of the fact that it has a half life much greater than its parent.  $^{237}\text{Np}$  emits an alpha (4.769 MeV) only 11 keV

Table 4. Relative hazard of transuranics in a representative reactor fuel element

Isotopes	Ci/Ci $^{90}\text{Sr}$	Ingestion DCFi/DCF $^{90}\text{Sr}$	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	1.70E-02
Cm-244	2.81E-03	14.1	4.00E-02

above  $^{234}\text{U}$  (4.758) and decays to  $^{233}\text{Pa}$  which emits a beta of average energy exactly the same as  $^{90}\text{Sr}$  (0.195 MeV).  $^{233}\text{U}$ , the daughter of  $^{233}\text{Pa}$ , emits alphas of 4.78 MeV (13.2 percent), only 2 keV from  $^{234}\text{U}$  and 4.82 MeV (84.4 percent) about 60 keV above  $^{234}\text{U}$  and 50 keV above  $^{237}\text{Np}$ .  $^{226}\text{Ra}$  emits an alpha (4.774) which is only 5 keV above  $^{237}\text{Np}$  and 16 keV above  $^{234}\text{U}$ . 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.

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

Review of surface water data for the last several years for Station 17 (East Fork Poplar Creek) and Outfall 304 (Bear Creek) shows that concentrations of all isotopes except for  $^{238}\text{U}$  and  $^{234}\text{U}$  were less than 1 percent of their DCGs. The concentrations of  $^{238}\text{U}$  and  $^{234}\text{U}$  were 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 drinking water limits. However, reported concentrations were often less than DLs or minimum detectable activities (MDAs). Therefore, MDAs must be lowered to provide acceptable confidence in the results (see Section 3).

The transuranium radioisotopes are expected to be less mobile than  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$ ,  $^3\text{H}$  or  $^{99}\text{Tc}$ . Therefore, although their hazard potential may be comparable to  $^{90}\text{Sr}$  and exceed  $^{90}\text{Sr}$  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 should continue using lower MDAs in order to confidently determine their contributions to the integrated dose limit (see Section 3).

### 2.3 Thorium Processing

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

Monitoring for these natural isotopes  $^{232}\text{Th}$ ,  $^{228}\text{Ra}$ ,  $^{228}\text{Th}$  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  $^{238}\text{U}$  decay chain specifically  $^{226}\text{Ra}$  and  $^{230}\text{Th}$ , 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.

### 3.0 BASIS OF DETECTION LIMITS

Analytical systems should have sufficient accuracy and precision to measure concentrations at reference levels or monitoring goals based on regulatory limits, background levels or for other uses. 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 DCGs. The detection limit for each isotope must be even lower since all isotopes are to be compared to their limits and summed to determine compliance. That is

$$\sum C_i/CL_i < 1$$

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

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

$$\sum C_i TF/CL_i < 1$$

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 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 (3 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 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 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 spect.
U-235	0.1	16.7	0.5	alpha spect.
U-234	0.1	16.7	0.5	alpha spect.
Am-241	0.1	16.7	0.5	alpha spect.
Np-237	0.1	16.7	0.8	alpha spect.
Pu-238	0.1	16.7	0.5	alpha spect.
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 spect.
Th-230	0.1	16.7	0.5	alpha spect.
Th-232	0.1	16.7	0.5	alpha spect.
H-3	0.05	1	400	liquid scint.
Tc-99	0.4	1	8	liquid scint.
Sr-90	0.1	1	15	gas flow prop.
Cs-137	0.9	16.7	6	gamma spect.

\* The MDAs also depends 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  $^{230}\text{Th}$  and  $^{232}\text{Th}$  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 U are for natural uranium which at 1 pCi/L would contain about 0.33 pCi/L of  $^{238}\text{U}$ , 0.33 pCi/L of  $^{234}\text{U}$  and about 0.014 pCi/L of  $^{235}\text{U}$ . Therefore, the MDAs necessary to determine background concentrations of natural U 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.

In addition, as discussed in Section 3 of the RMP, the MDAs necessary to determine weight percent  $^{235}\text{U}$  are less than 0.01 pCi/L (0.005 ppb) for  $^{235}\text{U}$  and about 0.1 pCi/L for  $^{234}\text{U}$  for low levels of depleted uranium.

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