



REGULATORY GUIDE

OFFICE OF STANDARDS DEVELOPMENT

REGULATORY GUIDE 4.14

MEASURING, EVALUATING, AND REPORTING RADIOACTIVITY IN RELEASES OF RADIOACTIVE MATERIALS IN LIQUID AND AIRBORNE EFFLUENTS FROM URANIUM MILLS

A. INTRODUCTION

Section 20.106, "Radioactivity in effluents to unrestricted areas," of 10 CFR Part 20, "Standards for Protection Against Radiation," provides that a licensee shall not release to an unrestricted area radioactive materials in concentrations that exceed limits specified in 10 CFR Part 20 or as otherwise authorized in a license issued by the Nuclear Regulatory Commission. Section 20.201, "Surveys," of 10 CFR Part 20 further requires that a licensee conduct surveys of concentrations of radioactive materials as necessary to demonstrate compliance with Commission regulations.

Section 20.401, "Records of surveys, radiation monitoring, and disposal," requires that records of surveys be maintained.

Section 40.65, "Effluent monitoring reporting requirements," of 10 CFR Part 40, "Licensing of Source Material," requires the submission of semi-annual reports to the Commission specifying the quantity of each of the principal radionuclides released to unrestricted areas and such other information as the Commission may require to estimate maximum potential annual radiation doses to the public resulting from effluent releases.

Paragraph (c) of Section 20.1, "Purpose," of 10 CFR Part 20 states that every reasonable effort should be made by licensees to maintain radiation exposure and releases of radioactive materials in effluents to unrestricted areas as far below the limits specified in Part 20 as is reasonably achievable.

This guide describes programs acceptable to the NRC staff for measuring, evaluating, and reporting releases of radioactive materials in liquid and airborne effluents from typical uranium mills. In some

cases, modifications of the described programs may be accepted or required by the NRC staff depending on individual site characteristics, plant design features, unique operations, or other factors. The need for modified programs will be determined by the NRC staff on a case-by-case basis.

B. DISCUSSION

Information on the radionuclides in liquid and airborne effluents from uranium mills, ore piles, and tailings is needed

1. For evaluation by the NRC staff of the environmental impact of radioactive materials in effluents, including estimates of the potential annual radiation doses to the public.

2. To ascertain whether regulatory requirements have been met and whether concentrations of radioactive materials in liquid and airborne effluents have been kept as low as is reasonably achievable.

3. For evaluation by the licensee and NRC staff of (1) the adequacy and performance of effluent controls and (2) the ore and tailings retention systems.

It is essential to have a degree of uniformity in the programs for measuring, evaluating, recording, and reporting data on radioactive material in effluents. This guide provides a uniform basis for comparing data from different sources and permitting the preparation of consistent summaries of data for use by the NRC staff as bases for assessing a licensee's effluent controls and the potential environmental impact of radioactive material in effluents.

This guide outlines general guidelines for acceptable effluent monitoring programs. However, these guidelines are not requirements. The licensing re-

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quirements are determined by the NRC staff on a case-by-case basis during individual licensing review. Individual applicants or licensees may propose alternatives for new or existing effluent monitoring programs that need not necessarily be consistent with this guide. The justification for such alternatives will be reviewed by the NRC staff, and the acceptability of proposed alternatives will be determined on a case-by-case basis during individual licensing reviews.

C. REGULATORY POSITION

1. METHODS OF SAMPLING AND ANALYSIS

Effluent monitoring is required to (1) demonstrate compliance with 10 CFR Part 20 and any special conditions of the license, (2) allow evaluation of the performance of retention systems and effluent controls, and (3) permit evaluation by the NRC staff of environmental impact and estimation of the potential annual radiation doses to the public. Because radiation dose is dependent on the radionuclides to which the individual is exposed, monitoring programs should provide accurate information on the specific radionuclides in airborne effluents and any liquid effluents from the plant, ore piles, and tailings retention system.

Methods of sampling and analysis of the radionuclides associated with uranium milling are discussed in sources listed in the bibliography. The listing of these documents does not constitute an endorsement by the NRC staff of all of the methods in all of the listings. Rather, these listings are provided as sources of information to aid the licensee in developing a program.

2. SAMPLING PROGRAM

2.1 Airborne Effluents

2.1.1 Stack Sampling

Effluents from each stack should be sampled at least semiannually during normal operations. The sampling should be adequate for determination of the release rates and concentrations of natural uranium for all stacks. The sampling of the yellow cake drier and packaging stack should also be adequate for the determination of release rates and concentrations of thorium-230 and radium-226.

2.1.2 Sampling at Site Boundary

Air particulate samples should be collected continuously at a minimum of three site boundary locations. The sampling should be adequate for the determination of concentrations of natural uranium, thorium-230, radium-226, and lead-210. Normally, filters for continuous, ambient air samples are changed at least weekly.

The sampling locations should be determined according to the specific site and milling operation. The following factors should be considered in determining the sampling locations: (1) average meteorological conditions (windspeed, wind direction, atmospheric stability), (2) prevailing wind direction, (3) site boundaries nearest to mill, ore piles, and tailings piles, (4) direction of nearest residence, and (5) location of estimated maximum concentrations of radioactive materials.

Samples should be collected continuously for at least one week per month, for the determination of the concentration of radon-222. The sampling locations should be the same as those for the continuous air particulate samples. Normally, sampling time for radon is 48 hours or less; therefore several samples per week will need to be analyzed for each sampling location.

2.2 Liquid Effluents

All liquid discharges to unrestricted areas should be sampled continuously. The samples should be adequate to determine concentrations and release rates of natural uranium, thorium-230, and radium-226.

Samples of groundwater should be collected at least quarterly from sampling wells located hydrologically downslope from the tailings retention system.

Samples should be collected at least quarterly from any surface seepage that reaches an unrestricted area and any natural body of water, such as a lake or creek, that crosses from the restricted area into an unrestricted area. (Surface seepage is defined as seepage from the tailings area that comes to the surface before it reaches the unrestricted area.)

Samples collected from groundwater, surface seepage, or natural bodies of water should be adequate for the determination of concentrations of natural uranium, thorium-230, and radium-226.

Any unusual releases that are not part of normal operations should be sampled. The sampling should be adequate to determine release rates and concentrations of natural uranium, thorium-230, and radium-226.

2.3 Quality of Samples

Provisions should be made to ensure that representative samples are obtained by use of proper sampling equipment, proper locations of sampling points, and proper sampling procedures (see bibliography).

Samples collected at the same location may be composited for analysis if they represent a sampling period of one calendar quarter or less. Samples

should not be composited (1) if they represent a sampling period of more than one calendar quarter, (2) if they are from different sampling locations, or (3) if the samples are to be analyzed for radon-222.

Samples collected for analysis of radon-222 should be analyzed quickly enough to minimize decay losses and allow for adequate precision and accuracy of results.

2.4 Alternative Sampling Programs

Applicants or licensees may propose alternatives to the sampling programs outlined in this regulatory guide. It is anticipated that programs that do not include continuous air samples at the site boundary will include more extensive stack sampling and more sampling locations than are described in this guide, as well as meteorological data and additional environmental monitoring requirements.

3. ANALYSIS OF SAMPLES

3.1 Air Samples

Stack effluent samples should be analyzed for natural uranium. Samples from the yellow cake drier and packaging stack should also be analyzed for thorium-230 and radium-226. The volume discharge rate of the stack effluents should be measured to the extent that it is necessary to estimate radionuclide release rates.

Air particulate samples collected at the site boundary should be analyzed for natural uranium, thorium-230, radium-226, and lead-210.

Air samples collected at the site boundary should be analyzed for radon-222. (Note: NRC regulations allow the analysis of radon daughters instead of radon-222. However, the NRC staff does not recommend this option because (1) techniques for long-term measurements of radon daughters at low levels are more difficult and (2) measurement of radon daughter concentrations near the site boundary instead of radon-222 would make estimates of dose to the public more difficult.)

These results should be used to determine the radionuclide release rates for the stacks and the radionuclide concentrations for the stacks and the site boundary.

3.2 Liquid Samples

Liquid samples should be analyzed for natural uranium, thorium-230, and radium-226.

The volumes of liquid discharges should be measured to the extent necessary to determine the radionuclide release rate.

These results should be used to determine the radionuclide release rate for liquid discharges and the radionuclide concentrations for liquid discharges, groundwater, surface seepage, and natural bodies of water.

3.3 Solubility of Radioactive Material

Table II of Appendix B to 10 CFR Part 20 lists separate values for soluble and insoluble radioactive materials. Therefore, both the soluble and insoluble portions of radionuclides in an effluent should always be analyzed. In order to determine compliance with 10 CFR Part 20, the licensee has two options: (1) the licensee may analyze all of a particular radionuclide in a sample and assume that it has the solubility corresponding to the lesser value in Table II of Appendix B to 10 CFR Part 20, or (2) the licensee may separate the soluble and insoluble portions in a sample, analyze each portion separately, and report each result separately, referring to the respective maximum permissible concentrations for soluble and insoluble materials.

3.4 Lower Limit of Detection

The lower limits of detection for analysis of air particulate samples collected at the site boundary should be 0.1% of the concentration limits listed in Table II of Appendix B to 10 CFR Part 20. For example, the lower limits of detection should be 5×10^{-11} microcuries per milliliter for natural uranium, 8×10^{-11} microcuries per milliliter for soluble thorium-230, 2×10^{-11} microcuries per milliliter for insoluble radium-226, and 4×10^{-11} microcuries per milliliter for soluble lead-210.

The lower limit of detection for analysis of radon-222 samples should be 3×10^{-11} microcuries per milliliter.

The lower limits of detection for stack effluent samples should be 10% of the 10 CFR Part 20, Appendix B, Table II concentration limits.

The lower limits of detection for liquid samples should be 1% of the concentration limits listed in Table II of Appendix B to 10 CFR Part 20 for natural uranium, thorium-230, and radium-226.

Obviously, if the actual concentrations of radionuclides being sampled are higher than the lower limits of detection indicated above, the sampling and analysis procedures need only be adequate to measure the actual concentrations.

An acceptable method for calculation of lower limits of detection is described in the appendix of this guide.

4. PRECISION AND ACCURACY OF RESULTS

4.1 Random Error

The random error associated with the analysis of samples representing concentrations above the lower limit of detection should be calculated. The calculation should take into account all significant random uncertainties, not merely counting error.

For samples representing concentrations below the lower limit of detection (see appendix), the licensee has two options: (1) the licensee may calculate the standard deviation associated with the analysis, or (2) the licensee may merely report the result as less than the lower limit of detection with no statement of uncertainty.

For effluents with concentrations at or below the concentrations listed in 10 CFR Part 20, Appendix B, Table II, the standard deviation estimated for random error should be less than both of the following: (1) 50% of the count and (2) 10% of the appropriate concentration listed in Appendix B, Table II. For effluents with concentrations greater than the concentrations listed in Appendix B, Table II, the standard deviation estimated for single counts should be less than 10% of the count.

4.2 Systematic Error

If the analyst estimates that systematic errors associated with the analysis are significant relative to the random error, the magnitude of the systematic error should be estimated.

4.3 Calibration

Individual written procedures should be prepared and used for specific methods of calibrating all sampling and measuring equipment, including ancillary equipment. The procedures should ensure that the equipment will operate with adequate accuracy and stability over the range of its intended use. Calibration procedures may be compilations of published standard practices, manufacturers' instructions that accompany purchased equipment, or procedures written in house. Calibration procedures should identify the specific equipment or group of instruments to which the procedures apply.

To the extent possible, calibrations of measuring equipment should be performed by using radioactive sources that have been calibrated by a measurement system traceable to the National Bureau of Standards' radiation measurements system.*

Calibrations should generally be performed at regular intervals. Frequency of calibration should be

*L. M. Cavallo et al., "Needs for Radioactivity Standards and Measurements in Different Fields," *Nuclear Instruments and Methods*, Vol. 112, pp. 5-18, 1973.

based on the stability of the system. If appropriate, equipment may be calibrated before and after use instead of at arbitrarily scheduled intervals. Equipment should be recalibrated or replaced whenever it is suspected of being out of adjustment, excessively worn, or otherwise damaged and not operating properly. Functional tests, i.e., routine checks performed to demonstrate that a given instrument is in working condition, may be performed using sources that are not calibrated by a system traceable to the National Bureau of Standards.

4.4 Quality of Results

A continuous program should be prepared and implemented for ensuring the quality of results and for keeping random and systematic uncertainties to a minimum. The procedures should ensure that the samples are not changed prior to analysis because of handling or because of their storage environment. Tests should be applied to analytical processes, including duplicate analysis of selected effluent samples and periodic cross-check analyses with independent laboratories.

5. REPORTING OF RESULTS

5.1 Sampling and Analysis Results

5.1.1 Air Samples

For each air sample, the following should be reported:

1. Location of sample.
2. Dates during which sample was collected.
3. For analyses indicating results above the lower limit of detection:
 - a. The concentrations of natural uranium, thorium-230, radium-226, and radon-222 for site boundary samples.
 - b. The concentration of natural uranium for stack effluent samples, plus the concentrations of thorium-230 and radium-226 for yellow cake drier and packaging stack effluent samples.
 - c. The percentage of the appropriate 10 CFR Part 20 Appendix B concentration limit.
 - d. The estimated release rate of natural uranium for stack effluent samples, plus the release rates of thorium-230 and radium-226 for yellow cake drier and packaging stack effluent samples.
4. For analyses indicating results below the lower limit of detection:

- a. The same information requested in (3) above or
- b. An indication that the results were below the lower limit of detection and the value of the lower limit of detection.

5.1.2 Liquid Samples

For each liquid sample, the following should be reported:

1. Location of sample.
2. Date of sample collection.
3. For analyses indicating results above the lower limit of detection:
 - a. The concentrations of natural uranium, thorium-230, and radium-226.
 - b. The percentage of the appropriate 10 CFR Part 20, Appendix B concentration limit.
 - c. For discharges to unrestricted areas, the release rates of natural uranium, thorium-230, and radium-226.
4. For analyses indicating results below the lower limit of detection:
 - a. The same information requested in 3 above or
 - b. An indication that the results were below the lower limit of detection and the value of the lower limit of detection.

5.1.3 Error Estimates

Results that are not reported as below the lower limit of detection should always include error estimates. The standard deviation associated with the random error of the analysis should be reported for each result. If significant, an estimate of the magnitude of the systematic error should also be reported.

Results reported as below the lower limit of detection need not include error estimates. However, the value of the lower limit of detection should be included.

5.2 Supplemental Information

The following information should be included in the first effluent monitoring report. Subsequent reports should include only changes in this information.

1. Description of sampling equipment.
2. Description of sampling procedures, including sampling times, rates, and volumes.
3. Description of analytical procedures.
4. Description of calculational methods.
5. Discussion of random and systematic error estimates, including methods of calculation and sources of systematic error.
6. Description of the calculation of the lower limit of detection.
7. Discussion of the program for ensuring the quality of results.
8. Description of calibration procedures.
9. Discussion of any unusual releases, including the circumstances of the release and any data available on the quantities of radionuclides released.

5.3 Units

Radionuclide quantities should be reported in curies. Radionuclide concentrations should be reported in microcuries per milliliter. (In the International System of Units, a curie equals 3.7×10^{10} becquerels, a microcurie equals 3.7×10^4 becquerels, and a milliliter equals 10^{-6} cubic meters.)

Standard deviations for random error should be reported in the same units as the result itself. Estimates of systematic error should be reported as a percentage of the result.

Note: The Commission has discontinued the use in 10 CFR Part 20 of the special curie definitions for natural uranium and natural thorium (39 FR 23990, June 28, 1974). Reports to the Commission should use units consistent with this change.

5.4 Significant Figures

Results should not be reported with excessive significant figures, such that they appear more precise than they actually are. The reported estimate of error should contain no more than two significant figures. The reported result itself should contain the same number of decimal places as the reported error.

5.5 Format

The term "not detected" or similar terms should never be used. Each reported result should be (1) a value and its associated standard deviation or (2) an indication that the result was below the lower limit of detection and the value of the lower limit of detection.

D. IMPLEMENTATION

The purpose of this section is to provide information to applicants and licensees regarding the NRC staff's plans for using this regulatory guide.

Except in those cases in which the applicant proposes an acceptable alternative method for complying with specified portions of the Commission's

regulations, the method described herein will be used in the evaluation of license applications docketed after February 15, 1978.

If an applicant wishes to use this regulatory guide in developing submittals for applications docketed on or before February 15, 1978, the pertinent portions of the application will be evaluated on the basis of this guide.

Appendix

LOWER LIMIT OF DETECTION

For the purposes of this guide, the Lower Limit of Detection (LLD) is defined as the smallest concentration of radioactive material sampled that has a 95% probability of being detected. (Radioactive material is "detected" if it yields an instrument response that leads the analyst to conclude that activity above the system background is present.)

For a particular measurement system (which may include radiochemical separation):

$$LLD = \frac{4.66 s_b}{3.7 \times 10^4 EVY \exp(-\lambda \Delta t)}$$

where

LLD is the lower limit of detection (microcuries per milliliter);

s_b is the standard deviation of the instrument background counting rate (counts per second);

3.7×10^4 is the number of disintegrations per second per microcurie;

E is the counting efficiency (counts per disintegration);

V is the sample volume (milliliters);

Y is the fractional radiochemical yield (when applicable);

λ is the radioactive decay constant for the particular radionuclide; and

Δt is the elapsed time between sample collection and counting.

The value of s_b used in the calculation of the LLD for a particular measurement system should be based on the actual observed variance of the instrument background counting rate rather than an unverified theoretically predicted variance.

Since the LLD is a function of sample volume, counting efficiency, radiochemical yield, etc., it may vary for different sampling and analysis procedures. Whenever there is a significant change in the parameters of the measurement system, the LLD should be recalculated.*

*For a more complete discussion of the LLD, see *HASL Procedures Manual*, John H. Harley, editor, USERDA, HASL-300 (revised annually) and Currie, L. A., "Limits for Qualitative Detection and Quantitative Determination—Application to Radiochemistry," *Anal. Chem.* 40, 1968, pp. 586-93.

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