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U.S. NUCLEAR REGULATORY COMMISSION

REGULATORY GUIDE

OFFICE OF NUCLEAR REGULATORY RESEARCH

REGULATORY GUIDE 4.16
(Task CE 401-4)

MONITORING AND REPORTING RADIOACTIVITY IN RELEASES OF RADIOACTIVE MATERIALS IN LIQUID AND GASEOUS EFFLUENTS FROM NUCLEAR FUEL PROCESSING AND FABRICATION PLANTS AND URANIUM HEXAFLUORIDE PRODUCTION PLANTS

A. INTRODUCTION

Section 70.59, "Effluent Monitoring Reporting Requirements," of 10 CFR Part 70, "Domestic Licensing of Special Nuclear Material," requires licensees authorized to possess and use special nuclear material for processing and fuel fabrication, scrap recovery, or conversion of uranium hexafluoride to submit semiannual reports to the NRC specifying the quantity of each of the principal radionuclides released to unrestricted areas and such other information as the NRC may require to estimate maximum potential annual radiation doses to the public resulting from effluent releases. Section 40.65, "Effluent Monitoring Reporting Requirements," of 10 CFR Part 40, "Domestic Licensing of Source Material," requires each licensee authorized to possess and use source material in the production of uranium hexafluoride to submit semiannual reports similar to those required by § 70.59.

Section 20.106, "Radioactivity in Effluents to Unrestricted Areas," of 10 CFR Part 20, "Standards for Protection Against Radiation," prohibits a licensee from releasing radioactive materials to an unrestricted area in concentrations that exceed the limits specified in 10 CFR Part 20 or that exceed the limits otherwise authorized in a license issued by the Nuclear Regulatory Commission (NRC). Section 20.201, "Surveys," of 10 CFR Part 20 requires that a licensee conduct surveys of concentrations of radioactive materials as necessary to demonstrate compliance with NRC regulations. Section 20.401, "Records of Surveys, Radiation Monitoring, and Disposal," requires that records of surveys be maintained for the period specified.

Paragraph (c) of § 20.1, "Purpose," of 10 CFR Part 20 states that every reasonable effort should be made by licensees to maintain radiation exposure and releases as far below the limits specified in 10 CFR Part 20 as is reasonably achievable. Sections 20.105, "Permissible Levels of Radiation in Unrestricted Areas," and 20.106, "Radioactivity in Effluents to Unrestricted Areas," of 10 CFR Part 20 require licensees engaged in uranium fuel cycle operations subject to the provisions of 40 CFR Part 190, "Environmental Radiation Protection Standards for Nuclear Power Operations," to comply with 40 CFR Part 190.

This guide provides methods acceptable to the NRC staff for developing effluent monitoring programs to be described in license applications and for monitoring and reporting effluent data by licensees. The guidance is applicable to nuclear fuel processing and fabrication plants and uranium hexafluoride production plants.

Information collection activities mentioned in this regulatory guide are contained as requirements in 10 CFR Parts 20, 40, or 70, which provide the regulatory basis for this guide. The information collection requirements in 10 CFR Parts 20, 40, and 70 have been cleared under OMB Clearance Nos. 3150-0014, 3150-0020, and 3150-0009, respectively.

B. DISCUSSION

Information on the identity, concentration, and quantity of radionuclides in liquid and gaseous effluents from nuclear fuel processing and fabrication and uranium hexafluoride production plants 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.

*The substantial number of changes in this revision has made it impractical to indicate the changes with lines in the margin.

2. To enable the NRC staff to ascertain whether regulatory requirements have been met and whether concentrations of radioactive material in liquid and gaseous effluents have been kept as low as is reasonably achievable (ALARA).

3. For evaluation by the licensee and the NRC staff of the adequacy and performance of effluent controls.

It is essential to have a degree of uniformity in the programs for monitoring and reporting data on radioactive material in effluents. This guide provides a basis for uniform reporting, for comparing data from different sources, and for permitting the preparation of consistent summaries 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.

C. REGULATORY POSITION

1. METHODS OF SAMPLING AND ANALYSIS

The sampling and analysis methods used in the effluent monitoring programs should be chosen to provide information on the quantity and concentration of radionuclides in gaseous and liquid effluents. The bibliography in this guide provides useful references on sampling, analysis, statistical analysis, and the preparation and maintenance of effluent monitoring programs.

2. SAMPLING PROGRAM

2.1 Gaseous Effluents

Gaseous effluents from all operations associated with the plant, including such nonprocessing areas as laboratories, experimental areas, storage areas, and fuel element assembly areas, should be sampled.

For gaseous effluents from process confinement systems and process areas where material is handled in dispersible form, a representative sample of the effluent from each stack, vent, or other point of release should be collected continually for subsequent determination of quantities and average concentrations of radionuclides released. This sampling should be conducted regardless of the concentrations of radioactive material in the effluent.

Other gaseous effluents should also be continually sampled unless the licensee has established, by periodic stack sampling or by other means, that radioactivity in the effluent is insignificant. In such cases, the effluents should be sampled at least quarterly. If periodic sampling is used in lieu of continual sampling, the licensee should show, as supplemental information, that the samples are representative of actual releases. For the purposes of this guide, a gaseous effluent release is significant if the concentration averaged over a calendar quarter is equal to 10% or more of the appropriate concentration listed in Table II of Appendix B to 10 CFR Part 20.

The sampling program should be sufficient to permit a determination of the quantities of radionuclides and the average concentration of radionuclides being discharged from the plant. The sampling rate at each release point should be such that a representative sample of the effluent is collected. The volumes of gaseous effluents should be reported so the NRC staff can calculate the quantities of radionuclides discharged.

2.2 Liquid Effluents

Representative samples should be collected at each liquid release point for the subsequent determination of the quantities and average concentrations of radionuclides discharged in any liquid effluents that could reach an unrestricted area, including discharges to a sanitary sewerage system. For continuous releases, representative samples should be continually collected at each release point. For batch releases, a representative sample of each batch should be collected.

For some liquid effluents, the licensee may establish, by periodic sampling or by other means, that radioactivity in the effluent is insignificant. In such cases, the effluents should be sampled at least quarterly. The licensee should show, as supplemental information, that these samples are representative of actual releases. For the purposes of this guide, a liquid effluent release is significant if the concentration averaged over a calendar quarter is equal to 10% or more of the appropriate concentration listed in Table II of Appendix B to 10 CFR Part 20.

The sampling program should be sufficient to permit a determination of the quantities of radionuclides and the average concentration of radionuclides being discharged from the plant. The sampling rate at each release point should be such that a representative sample of the effluent is collected. The volumes of liquid effluents should be reported so the NRC staff can calculate the quantities of radionuclides discharged.

2.3 Quality of Samples

Provisions should be made to ensure that representative samples are obtained by the use of appropriate sampling equipment, proper locations of sampling points, and proper procedures for collection and storage of samples.

Gaseous samples collected at the same location may be combined for analysis if they represent a sampling period of one week or less. Liquid samples collected at the same location may be combined if they represent a sampling period of one month or less. Samples should not be combined if they are from different locations.

3. ANALYSIS OF SAMPLES

3.1 Gaseous and Liquid Samples

For nuclear fuel processing and fabrication operations, 10 CFR Part 70 requires that gaseous samples be analyzed at least weekly and liquid samples at least monthly for gross alpha and beta activity. For uranium hexafluoride production (licensed under 10 CFR Part 40), gaseous samples should be analyzed at least weekly and liquid samples at least monthly for gross alpha activity. Liquid samples of effluents from uranium hexafluoride production plants should be analyzed periodically for thorium by gross beta activity or some other method to ensure compliance with 10 CFR Part 20.

Radionuclide analyses should be performed on selected samples unless (1) the gross alpha and gross beta activities are so low that individual radionuclides could not be present in concentrations greater than 10% of the quantities specified in Table II of Appendix B to 10 CFR Part 20 or (2) the radionuclide composition of the sample is known through such operational data as the composition of the feed material.

Examples of cases in which operational data may not be adequate for the determination of radionuclide composition are (1) plants processing uranium in which extraction, ammonium diuranate precipitation, ion exchange, or other separation processes could result in concentration of thorium isotopes (principally thorium-234), (2) plants in which uranium of varying enrichments is processed during the period of consideration, (3) plants processing plutonium in which significant variation in the plutonium-238/plutonium-239 ratio among batches of plutonium and the continuous ingrowth of americium-241 would preclude the use of feed material data in determining the radionuclide composition of effluents, and (4) uranium hexafluoride production plants in which evaluations based on feed materials show significant changes in the radionuclide ratio (e.g., uranium, radium, thorium).

Radionuclide analyses should be made more often (1) at the beginning of the monitoring program until a predictable radionuclide composition of effluents is established, (2) whenever there is a significant unexplained increase in gross radioactivity, or (3) whenever a process change or other circumstance might cause a significant variation in the radionuclide composition. Reports in which estimates of quantities of individual radionuclides are based on methods other than direct measurement should include an explanation and justification of how the result was obtained. The results of the analyses of the samples for each release point should be used to determine (1) the total gross alpha and gross beta activity (as applicable) discharged, (2) the average concentration of gross alpha and gross beta activity (as applicable) discharged, and (3) the total activity and average concentrations of each of the radionuclides discharged.

3.2 Lower Limit of Detection

The lower limit of detection for any analysis should be not more than 5% of the concentration limits listed in Table II of Appendix B to 10 CFR Part 20. For example, the lower limits of detection for soluble uranium-238 should be not more than 1.5×10^{-13} microcurie per milliliter for air and 2×10^{-6} microcurie per milliliter for liquid.

If the actual concentrations of radionuclides being sampled are known to be higher than the lower limits of detection indicated above, the sampling and analysis procedures need only be adequate to measure the actual concentrations. However, in such cases, the lower limit of detection should be low enough to accommodate fluctuations in the concentrations of the effluent and the uncertainty of the lower limit of detection.

An acceptable method for calculating lower limits of detection is described in Appendix A to this guide.

4. PRECISION AND ACCURACY OF RESULTS

4.1 Random Error

For each reported value, the standard deviation should be calculated for the random error associated with the analysis. The calculations should take into account all significant random uncertainties. The appropriate calculations will depend on whether the reported value is based on (1) a single count, (2) an average of counts from several samples, some of which may be below the lower limit of detection, or (3) indirect methods such as calculations based on gross alpha counts.

4.2 Calibration

Individual written procedures should be prepared and used for calibrating all sampling and measuring equipment, including ancillary equipment such as air-flow measurement instruments. 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 by the licensee. 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 radionuclide 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. The frequency of calibration should be based on the use and stability of the system. If appropriate, equipment may be calibrated by counting standardized radionuclide sources during use. Systems should be calibrated more frequently when counting at low radiation levels than when counting at levels that are statistically significant from background. 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.

4.3 Quality of Results

A program should be prepared and implemented for ensuring the quality of results and for keeping random and systematic uncertainties to a minimum. Regulatory Guide 4.15, "Quality Assurance for Radiological Monitoring Programs (Normal Operations)–Effluent Streams and the Environment," provides useful guidance on quality assurance.

Procedures should be established to ensure that the samples are not affected by improper handling or storage prior to analysis. For example, liquid samples may have to be chemically treated to prevent losses to the walls of storage containers, and samples containing solids either should be made homogeneous or the liquid and solid portions should be analyzed and reported separately.

Tests should be applied to analytical processes, including duplicate analyses of selected effluent samples and periodic cross-check analyses with independent laboratories.

Because much of the data to be reported may be based on gross radioactivity measurements, the program should include periodic tests to ensure that such measurements represent actual quantities of individual radionuclides in samples. For example, in plants handling uranium, a chemical analysis for uranium should be performed at least quarterly on selected samples for comparison with the gross radioactivity analyses.

5. REPORTING RESULTS

5.1 Sampling and Analysis Results

All data should be summarized on a semiannual basis. For each release point, the following should be reported, as appropriate (see Appendix B to this guide for illustrative formats):

1. Type of sample (gaseous or liquid),

2. Sampling location,

3. Dates during which samples were collected,

4. The quantities of gross alpha activity, gross beta activity, and each radionuclide released,

5. The average concentrations of gross alpha activity, gross beta activity, and each radionuclide released,

6. The percentage of the appropriate concentrations listed in Table II of Appendix B to 10 CFR Part 20.

The following summary should be included in each report:

1. For all gaseous releases, the total quantities of gross alpha activity, gross beta activity (if appropriate), and each radionuclide,

2. For all liquid releases, the total quantities of gross alpha activity, gross beta activity (if appropriate), and each radionuclide,

3. For the gaseous sample observed to contain the highest concentration of radioactivity for the semiannual period, the concentrations of gross alpha activity, gross beta activity (if appropriate), and each radionuclide, along with the percentages of the appropriate concentrations listed in Table II of Appendix B to 10 CFR Part 20,

4. For the liquid sample observed to contain the highest average concentration of radioactivity for the semiannual period, the concentrations of gross alpha activity, gross beta activity (if appropriate), and each radionuclide, along with the percentages of the appropriate concentrations listed in Table II of Appendix B to 10 CFR Part 20.

If the highest concentrations in paragraphs 3 and 4 are less than 10% of those listed, the summary should indicate that the result was below the stated appropriate value.

5.2 Error Estimates

Reported results should include error estimates. The standard deviation representing the random error of the analysis should be reported for each result. An estimate of the magnitude of the systematic error should be reported separately. 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.

5.3 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, frequencies, rates, and volumes,

3. Description of analytical procedures,

4. Description of calculational methods, for example, calculation of radionuclide quantities using gross radioactivity measurements,

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,

10. The basis for any determination that a stack or liquid release point need not be continuously sampled.

5.4 Units

Radionuclide quantities should be reported in curies or multiples of curies as defined in § 20.5 of 10 CFR Part 20. Radionuclide concentrations should be reported in microcuries per milliliter.

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

5.5 Significant Figures

Results should not be reported with excessive significant figures so that they appear more precise than they actually are. The reported estimate of error may contain one significant figure. The reported result should contain the same number of decimal places as the reported error.

5.6 Format

A sample format is illustrated in Appendix B.

The term "not detected" or similar terms should not 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 stated 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 an applicant or licensee proposes an acceptable alternative method for monitoring or reporting radioactivity in effluents, the methods presented in this guide will be used in the evaluation of applications and licensee reports after December 1985.

BIBLIOGRAPHY

American National Standards Institute, Inc., "Guide to Sampling Airborne Radioactive Materials in Nuclear Facilities," ANSI N13.1-1982, New York, 1982.

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National Council on Radiation Protection and Measurements, "A Handbook of Radioactivity Measurements Procedures," NCRP Report No. 58, Washington, DC, 1978.

APPENDIX A

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):

$$LLD = \frac{2.71 + 4.66S_b}{3.7 \times 10^4 \, EVY \exp(-\lambda \Delta t)}$$

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 the midpoint of sample collection and the time of 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 on 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 *EML Procedures Manual*, H. L. Volchok and G. dePlanque, editors, U.S. Department of Energy (revised annually); and L. A. Currie, "Limits for Qualitative Detection and Quantitative Determination– Application to Radiochemistry," *Analytical Chemistry*, Vol. 40, pp. 586-593 (Table II), 1968.

APPENDIX B

Example of Format For Reporting Effluent Data[1]

1. CONTINUOUSLY SAMPLED STACKS

For each release point, report the following information:

a. Reporting period
b. Stack location (process or area)
c. Stack flow rate (m^3/sec) or total stack flow (m^3) (if stack is not in continuous use)

Radioactivity[2] (Ci)

Gross alpha
Gross beta

Radionuclide[2]	Concentration (μ Ci/ml)	Error Estimate[3] (± μ Ci/ml)	LLD[4] (μ Ci/ml)	Quantity Released (Ci)	% MPC
U-234					
U-235					
U-238					

[1] This table is for illustration only and is not a complete listing of data to be reported. Supplemental and explanatory information should also be submitted. (See footnote 2 and text of guide, for instance, sections 2.1 and 2.2.)

[2] This list of radioactivity and radionuclides is typical for uranium fuel fabrication plants. It is not complete for all licensees, nor will all licensees need to report the radioactivity or radionuclides shown. (For example, some licensees may need to report natural uranium, isotopes of plutonium, etc., and some licensees may not need to report gross beta activity.)

[3] Estimates of random errors should be calculated at the 95% confidence interval. Significant systematic error should be reported, if appropriate, in supplemental information.

[4] Calculation of lower limits of detection (LLD) should be shown in supplemental information.

2. OTHER SAMPLED STACKS[5]

For each release point, report the following information:

a. Date(s) sampled
b. Stack location
c. Stack total flow (m^3)

Radioactivity[2] (Ci)

Gross alpha
Gross beta

Radionuclide[2]	Concentration (μ Ci/ml)	Error Estimate[3] (± μ Ci/ml)	LLD[4] (μ Ci/ml)	Quantity Released (Ci)	% MPC

3. LIQUID SAMPLES

For each release point, report the following information:

a. Reporting period or date(s) sampled
b. Location of sample collection
c. Total liquid flow
d. Batch or continuous sample
e. Dilution flow (if not included in total flow)

Radioactivity[2] (Ci)

Gross alpha
Gross beta

Radionuclide[2]	Concentration (μ Ci/ml)	Error Estimate[3] (± μ Ci/ml)	LLD[4] (μ Ci/ml)	Quantity Released (Ci)	% MPC
U-234 (dissolved)					
U-234 (suspended)					
U-235 (dissolved)					
U-235 (suspended)					
U-238 (dissolved)					
U-238 (suspended)					

[5]This section is intended to cover stacks or vents not routinely sampled (see Section 2.1). (For example, some stacks or vents may need to be sampled periodically to verify that radioactive effluents are insignificant or that the systems are used only intermittently.)

VALUE/IMPACT STATEMENT

A draft value/impact was published with the proposed Revision 1 to Regulatory Guide 4.16 (Task CE 401-4) when the draft guide was published for public comment in September 1984. No changes are necessary, so a separate value/impact statement for the final guide has not been prepared. A copy of the draft value/impact is available for inspection and copying for a fee at the Commission's Public Document Room at 1717 H Street NW., Washington, DC.

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