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REPORT

**EFFLUENT MONITORING AND EVALUATION--
A POWER REACTOR DESIGN GUIDE**

I. C. ROBERTS

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A POWER REACTOR DESIGN GUIDE

By

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EFFLUENT MONITORING AND EVALUATION--A POWER REACTOR DESIGN GUIDE

INTRODUCTION

The successful operation of a power reactor facility, or almost any facility utilizing radioactive materials, requires that the facility operator be aware of the consequences of releasing radioactive materials to the environment and that he exercise appropriate control over such releases. The big question is what are the consequences and how can the operator become informed about them. If he attempts to study the subject, he finds it to be a complexity of suggested and traditional radiation protection practices, governmental regulations, and poorly defined liability and public relations requirements. This "Guide" attempts to clarify the subject by identifying and quantifying the many factors involved.

Because of the rapid growth of the nuclear power industry an urgent need exists for a basic clarification of effluent and environmental monitoring requirements has arisen. The scope of this report is directed toward power reactor practices and, particularly, to conditions existing during normal operation of power reactors. Evaluation of accident conditions is not included because it is currently receiving much special research attention.

The material in this document and its manner of presentation has been selected to meet the needs of readers who wish to understand the basic requirements of monitoring and, consequently, will not always meet the needs of readers who want to be given "cook-book" procedures. The justification for

this approach is based on the belief that progress will best be served if power reactor designers and operators are informed about basic monitoring requirements and allowed to use their own ingenuity in developing systems to fulfill these requirements.

SUMMARY

The adequacy and effectiveness of the radioactive waste control practices employed at a power reactor plant must be assessed by monitoring the effluent streams and, in some cases, by monitoring environmental media. The principal reason for controlling the release of radioactive wastes is the minimization of radiation exposure of persons who inhabit the environs. Therefore, the monitoring information obtained must relate to radiation exposure and, preferably, to the radiation doses actually delivered. This can be accomplished most effectively by relating release data to "safety limits" at each power reactor plant. Such a limit represents a condition quite different than is encountered during normal operations. Exceeding the limit by a moderate amount does not, in itself, result in serious consequences but clearly indicates a significant departure from normal conditions.

The safety limit for the discharge of radioactive wastes is that concentration (or quantity) which, if continued, could cause the radiation doses delivered to persons in the environs to exceed permissible levels.

The words "if continued" are important because radiation doses are a function of time; and when doses are referred to, some period of time must be specified.

Another definition of a safety limit for radioactive waste discharge is that it is the Curie quantity or concentration of wastes which, if discharged, could result in a maximum permissible concentration of a particular radionuclide or combination of radionuclides in the environs. The definition of maximum permissible concentration (MPC), in turn, is that concentration of radionuclide(s) which, if continuously taken into the body of a person, could result in a maximum permissible dose being delivered to one or more organs, or to the whole body of that person. To make sure that concentrations of radionuclides in the environs do not exceed maximum permissible levels, some control must be exercised over the release of the wastes producing the environmental concentrations. To estimate permissible release rates (or quantities), many complex variables, most of which tend to dilute and some to concentrate the radionuclides must be considered. For effluents, those permissible release rates are the safety limits mentioned above.

If the safety limits are not to be exceeded, a warning that the limits are being approached is needed. Instantaneous or short term release rate limits are commonly employed for this purpose. These limits are called "release guides" or "operating standards". Under the current regulatory

process, the plant operator states, as a part of his application for license, the operating guides within which he will operate. When the license to operate is approved, operation of the plant within the approved guides becomes a condition of compliance with regulatory requirements.

Actually, operating guides define the limit of normal operating conditions. If they are exceeded, special action is required to determine that the principal regulatory requirement (annual average concentration) and primary radiation protection requirements (dose per year) are not also being exceeded. To do this, actual measurement of dose rates at strategic locations in the environs are required. Thus, the plant designer can either design the waste containment and treatment systems with sufficient capability for controlling releases of radionuclides to the tenths or hundredths of permissible rates and, thereby, virtually eliminating the need for an environmental monitoring system, or he can release radionuclides at rates that could conceivably cause dose rates near permissible levels.

The establishment of safety limits requires the application of a dilution factor between the point of discharge and the point of exposure. The dilution results from the mixing and exchange that takes place between the released materials and the receiving media. Many significant contributions to the understanding of diffusion phenomena have been made since the turn of the century; however, presently, the mathematical models of

diffusion most applicable to power reactor practice are semiempirical and must be fitted to the particular conditions at each installation. In cases where there are not enough environmental data to reliably "fit" these semiempirical equations, certain conservative estimates, based on experience gained at other locations, of environmental conditions have to be made.

Various chemical elements are concentrated in the environment, particularly by the biota. Therefore, after dilution factors are calculated for particular radionuclides to be released, the possibility of their being reconcentrated must also be explored before a safety limit can be established. For example, the concentration of radioiodine in the stack-grass-cow-milk-man exposure pathway causes the release of radioiodine to be of principal interest in power reactor waste disposal practice.

If it is possible for liquid radioactive wastes to be released in amounts approaching the safety limit established at a particular plant, then those wastes must either be (1) retained and measured prior to release, or (2) monitored as they are released.

Even if all liquid wastes are retained and analyzed before being released, it is still possible that some unforeseeable equipment or procedural failure can cause contaminated waste to be released. Therefore, a warning monitor on all liquid effluent streams is needed, with a possible exception of the main condenser coolant discharge of plants using very large quantities of coolant water. An event that would

contaminate such a large volume of water to a significant level would surely be observed by process stream monitors as well as by area monitors within the plant.

Release rate of radionuclides via a large effluent stream is more reliably determined by measuring the additives and calculating the diluted concentration than by measuring the diluted stream directly. Effluent monitoring in such cases should be limited to alarm-type devices.

The routine identification of individual radionuclides is not required. Routine measurement of total beta-gamma activity meets all effluent monitoring requirements if supplemented by an initial identification of the nuclides in each waste stream and by periodic confirmation that the composition has not changed significantly.

The release of gaseous wastes during the normal operation of a power reactor can possibly involve, significantly, two exposure pathways, viz. (1) whole body exposure from the release of noble gases, and (2) iodine reconcentration in the air-grass-cow-milk-man pathway. The release rates of the noble gases and radioiodine must, therefore, be monitored. The accuracy, required of the data, increases as the possible release rates approach permissible limits. Plants that anticipate release rate approaching 10% of the safety limit should employ continuous monitors, accurately calibrated and frequently maintained. All plants should have monitors that sound a warning when the instantaneous discharge rate approaches a level that could cause the permissible annual average release rate to be exceeded.

Every plant releasing radioactive

materials should be able to reliably estimate the resulting radiation doses received by the people inhabiting the plant environs. Because of the low rates and concentrations at which power reactor plants release radioactive wastes, the resulting concentrations in environmental media are too small to be measured by practical means and have no radiation exposure significance. However, the release of radioiodine is an exception because it very effectively concentrates in the environmental pathway leading from air through cow's milk to man. Consequently, there should be a milk sampling program in place around plants that release an average of 10 mCi/week or more of I^{131} (no current reactor releases reach that level) because 10 mCi/week, released from a 250-foot high stack, will produce I^{131} concentration in milk of about 2pCi/l. The maximum detection limit for I^{131} in milk is about 10 pCi/l if the iodine is concentrated by ion exchange. Therefore, most of the milk samples analyzed by a plant normally discharging an average of 10 mCi/week of I^{131} will have iodine concentrations below detectable limits. The plant will be prepared to detect any increase due to an abnormally high release rate, however.

With the release of noble gases, it is possible that people in the environs may receive external whole body radiation exposures. Measures controlling release rates of these gases are developed as an integral part of the reactor plant design. Control effectiveness should be checked by trying to measure, possibly by dosimeters, the actual dose

delivered at several points in the environs. Past experience of operating power reactors shows that the dose delivered via this route is too small to be measured.* The potential benefits in continuing such external dose measurements, after the design estimates are confirmed, are questionable. However, the ease and slight expense with which the monitoring can be performed are the chief advantages for continuing. Another advantage is the public relations value to a monitoring record that shows the very low doses that have resulted if, in fact, they are even measurable.

Much of the environmental monitoring performed from currently operating power reactors is done for public relations reasons. The negative results obtained, when environmental media are monitored, assures people using these media that their health is being adequately protected. The extent of sampling done depends largely on the public relations policies of the companies and organizations involved.

The experience gained in planning and executing a preoperational environmental monitoring can be directly useful when operations begin, but the data collected under such a program have little value. Under the conditions

* An exception to this statement occurred at Humboldt Bay where measurable off-site doses from noble gases were observed. Stack release rates during this period increased to the noble gas limit of 50,000 μ Ci/sec because of defective fuel cladding. The maximum off-site dose was such that it would have been one third of the annual limit of 500 mrem above background if this release rate were sustained for twelve months.

of weapons testing fallout, which have prevailed during the past decade, the levels of "background" activity vary so much with time that it is not possible to establish a preoperational baseline for comparing postoperational levels.

Current background measurements, made in the same geographical area as the reactor plant but far enough away that the plant cannot possibly affect the measurements, provide the most useful comparative data. Such data are collected by the U.S. Public Health Service Radiological Surveillance Network and published under the title, Radiological Health Data.

EFFLUENT MONITORING

Objectives

Effluent monitoring fulfills two operating requirements at a power reactor plant. It provides notification when safety limits are being approached and also provides certain useful data.

Using present practices as a reference, there are seven reasons, or objectives, for the collection of effluent monitoring data:

- To provide feedback for plant operation
- To estimate the radiation dose delivered to persons inhabiting the environs of a nuclear power plant
- To demonstrate compliance with governmental regulations specifying waste release limits
- To provide evidence as possible testimony in defense of a personal damage claim
- To promote good public relations
- To provide research data in the

study of atmospheric dispersion and dilution during various conditions at a given site

- To provide environmental exposure data during accident conditions.

Radiation Dose Estimation

The primary standard of radiation protection is the radiation dose received. Maximum permissible doses of radiation have been recommended by competent national (National Committee on Radiation Protection, Federal Radiation Council) and international (International Commission on Radiological Protection) organizations. The operator of every nuclear facility should assess the doses delivered to persons in the environs of his plant (from the operation of his plant) and compare these against the recommended limits. Actually, the doses are very low, much smaller than recommended limits, and cannot be measured directly. Therefore, they must be estimated from effluent monitoring data. The dose delivered to persons in the environs of a nuclear plant can be modelled as follows:

$$D = (Q - L) \text{ D.R. } \times F \quad (1)$$

where

D = dose rate [rads (or rems)* per unit of time]

Q = release rate (Curies per unit of time)

* The term "rad" is the unit of absorbed dose and is defined as the absorption of 100 ergs of absorbing material. The "rem" is a dose equivalent term and is numerically equal to the dose in rads multiplied by appropriate modifying factors; that is, a linear-energy transfer-dependent factor and a distribution factor for internally deposited radionuclides.

L = loss rate between point of measurement and point of exposure such as radioactive decay, and immobilization of radionuclides in the environment. (Curies per unit of time).

D.R. = dilution rate between point of measurement and point of exposure in cubic meters per unit time.

F. = concentration to dose rate factor [rads (or rems) per Curie per meter⁻³]

The release rate can either be measured directly, if the plant is operating; or assumed, if the plant is in the design stage. The important losses such as radioactive decay, and settling of particulates must be estimated. Dilution (or dispersion) factors can be estimated by using semiempirical equations, but these estimates require confirmation. The concentration to dose rate conversion factor is supplied by ICRP Committee II report and the FRC report as explained below.

The release of radioactive materials can expose persons in the environs to sources of radiation external to their bodies. In addition, ingestion and inhalation of radioactive materials, produces internal exposure. The ICRP Committee II and the FRC have calculated the concentrations of radionuclides in air and water that can result in maximum permissible doses to people. These concentrations are termed the "maximum permissible concentration" by ICRP while the FRC uses the term "Radiation Protection Guide." Since the relationship between concentration and

dose can be assumed to be linear, and the maximum permissible doses are given by ICRP and FRC, the doses delivered by concentrations different than the maximum permissible concentrations can be estimated by using sample proportions. For example, FRC Report No. 2 states that a continuous intake of 100 pCi/day of I¹³¹ will result in a dose to the thyroid of an infant of 0.5 rem/yr, the dose recommended as a "Radiation Protection Guide." It follows, therefore, that a smaller daily intake will deliver a proportionately smaller dose e.g., an intake of 50 pCi/day delivers a dose of 0.25 rem/yr.

It should be noted that the release rate term in the above model is expressed in units of quantity not concentration. The expansion of an initial quantity of waste released into an "infinite" volume such as the atmosphere or a very large water body is unaffected by the original concentration (i.e. quantity/volume of the waste).

The total radiation dose delivered by a released waste is a composite of the dose delivered by each radionuclide in that waste. The relative importance of each radionuclide's contribution depends upon factors described in Equation 1. Considerable difference exists in the hazard potential of the various radionuclides in any given effluent and, in theory, each radionuclide should be investigated if the total dose is to be accurately estimated. In almost all cases most of the total dose is contributed by one or two radionuclides and the contributions of all other radioconstituents are insignificant.

Compliance with Regulatory Requirements

A power reactor operator must comply with the Federal regulations published as 10 CFR Part 20 and with any additional requirements imposed by the USAEC Division of Licensing and Regulation as a condition of approval to operate. Other Federal agencies, such as the Federal Radiation Council and the U.S. Public Health Service, have a strong interest in the radiation protection practices of nuclear facilities, but exercise no direct regulatory control over them. Various state and local agencies also have an interest in radiation protection but are excluded from exercising control over reactor facilities except by agreement with the USAEC. States' interest is perhaps best reflected in a Memorandum Understanding signed by the USAEC and the State of New York. This document was published in the Federal Register on May 20, 1965. It states that the authority of the state to impose regulatory requirements on USAEC licensees that are more restrictive than the AEC's own requirements is not recognized. If a state (including a nonagreement state) believes that additional restrictions should be imposed on an AEC licensee's operation, they are requested to deal with the AEC instead of the licensee. The Memorandum of Understanding also recognizes that a state government may make appropriate surveys, sample effluents, and examine certain records mentioned by AEC licensees. Pertinent regulations are summarized and discussed in Appendix A. The functions and responsibilities of the various regulatory agencies are also discussed in Appendix A.

The regulations published as 10 CFR, Part 20 consist of maximum concentrations of stated radionuclides and mixtures of radionuclides. These are considered minimum requirements. The operator of a licensed facility is required to prove that his operations have complied with these limits by submitting evidence in the form of effluent and process monitoring data.

Release limits and, in some cases, permissible environmental concentrations for particular radionuclides in addition to those listed in 10 CFR, Part 20 are imposed upon reactor facilities by the Division of License and Regulation as a conditions of operating approval. In the past, these special requirements have usually involved the release of radioiodine and noble gases.

The operating procedures for limiting offsite exposures around a reactor facility are evaluated three times by the AEC regulatory divisions before approval to operate is given. They are reviewed first by the Reactor Licensing Staff which uses the prevailing standard that off-site exposures should not exceed 0.5 rem/yr above background. The Staff considers the site meteorological characteristics and the nature of the effluents, and determines permissible maximum instantaneous release rates and cumulative releases.

As a power reactor begins operating, the operator has to prove to the appropriate regional officer of the Division of Compliance that his operations comply with pertinent federal regulations. The Division of Compliance does not require specific data to support the operator's claim of compliance with regulations. Whatever data the operator chooses to submit are reviewed

and the pertinence and accuracy of the data are evaluated during a plant inspection.

The operating reactor plant is visited (inspected) several (~ 3 to 10) times a year by representatives of the Division of Compliance as a part of that organization's reactor inspection program.

Legal Evidence in Personal Damage Claims

Very little experience exists in the litigation of personal injury and damage claims involving the release of radioactive materials. Therefore, the evidential value of health physics data in future claims can only be conjectured. There can be little doubt, however, that health physics data would be used--and probably extensively--if available. The collection of such data is expensive, and the manager of each nuclear plant must decide how much data will be collected.

The exact value of data is yet undetermined, but a lack of data could be construed as negligence. It is likely that the operator of the plant will have to provide sufficient evidence to disprove the allegation.

The reliability and interpretation of the data collected would be principal factors in the admissibility of health physics data as evidence. Shapar (1961) cites the following court statement:

"Just when a scientific principle or discovery crosses the line between the experimental and demonstrable stages is difficult to define. Somewhere in this twilight zone the evidential force of the principle must be recognized and while courts will go a long way in admitting expert testimony deduced from a well-

recognized scientific principle or discovery, the thing from which the deduction is made be sufficiently established to have gained general acceptance in the particular field in which it belong".*

A prime requirement of data collected as possible evidence is that they be collected systematically and represent as closely as possible the true conditions that existed at all times. Absolute precision is impossible for technical reasons, and so any adapted monitoring scheme will have to be a compromise of the various factors involved.

The first element of a negligence case is whether reasonable care has been exercised. Reasonable care could mean that the most precise and reliable instruments and techniques have been used to assure minimum radiation exposures. A very important point here is that compliance with pertinent governmental regulations may not constitute reasonable care any more than compliance with a speed limit denotes the exercise of reasonable care in driving a vehicle. However, as a result of the extreme attention given to radiation safety by governmental agencies, it would seem to be highly unlikely that negligence could remain undetected and/or persist at a power reactor plant.

Public Relations

The power and effect of public opinion is well recognized and has been felt in every segment of the modern industrial society. The atomic energy industry has received perhaps more than its normal quota of public

* *FRY versus United States*, 293 Fed. 1013, 1014 (D.C. Cir. 1923)

attention due to the anxieties aroused by atomic weapons. Consequently, the industry has tended to be ultrasensitive to public opinion and oftentimes has adopted extra conservative design criteria and practices to avoid adverse public opinion.

Despite the strong influence that public opinion has had on the industry, very little attention has been given to analysis of the factors influencing that opinion. From experience has come an intuitive awareness of what is important and unimportant to the public but an organized system for evaluating and improving this wisdom has not yet evolved.

There seem to be two conflicting philosophies on environmental radiation protection at nuclear power plants. One school of thought advocates maximum demonstration of concern for the safety of public in every way possible; e.g., information meetings, publication of monitoring results even to the extent of having monitoring personnel and vehicles seen as often as possible. Others maintain that an over concern for safety by management of a nuclear power plant only increases concern and suspicion in the public. This second group points out also that proof of absolute safety is not possible, and that any data collected to demonstrate safety can be impugned. Therefore, defense of the data and their meaning puts the well intentioned plant manager in an undesirable position.

In matters involving the complex emotional characteristics of human beings, it may be a long time before it is possible to isolate and evalu-

ate the public relations factors extant in a community as it is perturbed by the introduction of a power reactor.

Requirements

Two of the monitoring objectives described previously, viz., radiation protection and compliance with governmental regulations, comprise the principal monitoring requirements at a power reactor facility. Effluent monitoring information is required to calculate the radiation dose delivered to persons in the environs and to demonstrate compliance with regulatory requirements. In large part, the requirements of the two objectives are identical because radiation protection is the essence of the regulations.

In theory it is necessary to identify and quantify each released radionuclide so that the dose contribution of each can be estimated. (A discussion of the radionuclide composition of current reactor systems is attached as Appendix B, "Identification of Radionuclide Composition of Power Reactor Wastes.") In current reactor practice, only the radioiodine and inert gases, released as gaseous wastes, require special attention. However, some question does exist as to the future long-term importance of tritium released in liquid wastes. A short discussion of the hazard potential of the radioiodines and inert radiogases is appended.

The inevitable question is how precisely the concentrations or rate of release of radioactive materials in effluent streams must be measured. It can be stated that, in general, the closer the measured value approaches some limit, such as an operating limit,

the more precise the data must be. If the data represent normally distributed population, the error in average value measured (S.E.) can be expressed as

$$\text{S.E.} = \frac{\sigma}{\sqrt{N}}, \quad (2)$$

where

σ is the standard deviation of the data and N is the number of samples taken. This method of estimating standard error can be useful even if the population sampled is not normally distributed.

When monitoring data are compared to a maximum permissible value, it is necessary to know the probability that the mean value, m , of the data plus the standard error (S.E.) is less than the limit. That is,

$$m + \text{S.E.} < \text{MPC}. \quad (3)$$

For limits to be meaningful, the probability that they have been exceeded must be kept very low, say 1 in 1000. This can be expressed statistically by using a 3σ for the numerator in Equation 2, viz.,

$$\text{S.E.} = \frac{3\sigma}{\sqrt{N}}, \quad (4)$$

with a 0.999 probability that the S.E. is never greater than calculated.

It is apparent from Equations 2 and 4 that the S.E. decreases as the square of the number of sampling events increases. A decrease in the S.E. is possible if σ can be reduced. The variance inherent in the population being sampled cannot be changed, of course, but the precision in effluent sampling data can be improved not only by increasing the sampling frequency but also by improving the

precision of the analytical procedures used.

Actually, the foregoing theory is infrequently applied because the release rates of radioactive materials in effluent streams are so much lower than safety limits, thus the precision requirements can be set intuitively. For example, the mean concentration of liquid waste prior to dilution at the Rowe reactor plant from 1961 through 1963 was reported as 2.4×10^{-6} $\mu\text{Ci/ml}$, with an available dilution factor of 4700. The resultant average concentration of radioactivity in the diluted effluent can be calculated to be 5×10^{-10} $\mu\text{Ci/ml}$. If this concentration is compared to the most restrictive MPC in 10 CFR Part 20, 10^{-7} $\mu\text{Ci/ml}$ for mixtures of unidentified radionuclides, a standard error of $\pm 100\%$ is tolerable.

While the determination of the mean concentration range of radioactive materials in a medium, especially an effluent stream has been discussed, some applications of monitoring data probably do, in fact, require quantitative values. For example, if monitoring data are to be used in the computation of doses received by persons in the environs, a quantitative expression of the amount of released radioactive material is required. Strictly speaking, however, the quantitative expression of the average annual concentrations of radioactive materials in an effluent stream is not required to comply with the regulations which state that a maximum annual average concentration shall not be exceeded. All that is needed, actually, is reliable proof that the

limiting average has not been exceeded. Determination of the actual concentration is supererogatory. A monitoring system providing assurance that the average concentration radioactive materials released has been within limits is desirable. Such a system has not yet been developed within the atomic energy industry. A precedent does exist in the British coal industry where a maximum concentration of dust particles in the air (which the miners breathe) is established by regulation. A simple sequential procedure, testing whether average conditions achieve the prescribed standard, is used. (See Appendix C)

Most of the liquid radioactive wastes generated by power reactors are retained temporarily in tanks. The contents of the tanks can be analyzed before being released in a controlled manner to large volume streams of virtually nonradioactive diluting medium such as the main condenser coolant water. As experience is gained, the extra degree of control afforded by this system may prove unwarranted in view of the concomitant capital investment in tanks and associated piping, and the operating expense in sampling the tanks. Also, the philosophy of temporarily retaining all wastes may prove to be impractical and unnecessary in certain types of reactors. In these cases the use of continuous monitors or of a well-developed sampling program will be mandatory.

Sampling hold-up tanks involves no particular statistical problem because the gases and liquids contained in the tanks are homogeneous media, and a

single sample is indicative of the entire contents. However, if a tank contains particles of solid material which have settled into a sludge on the bottom of the tank and which will remain with the bulk media when it is discharged, the sludge should be mixed throughout the tank contents before sampling.

The continuous flow of liquid waste material in an industrial plant characteristically shows irregular swings around a mean value with a period varying from minutes to days according to the nature of the plant. In addition, the mean value, itself, swings about in an irregular manner over much longer periods of time. Somewhat the same pattern could be expected in a continuous waste effluent from a power reactor. Any attempt to monitor such a stream quantitatively must reckon with considerable radioactivity from radio-nuclides with very short half-lives. As described in Appendix B, these might be products of fission or activation. As such, the variations in their intensity is primarily a function of neutron flux level, i.e., reactor power level. Changes in leakage rates, removal efficiencies of waste treatment processes, number and size of fuel cladding are typical of other factors that might vary the characteristics of a continuously flowing stream.

Sampling a continuously flowing stream for a characteristic having a variable mean such as reactor plant effluent is fraught with problems. The most serious of these is how to interpret the data collected, e.g., are observed fluctuations indicative of significant trends? There is also

the problem of establishing a sampling frequency. The use of a proportional sampling device may solve the frequency problem, however. Many of these problems can be avoided if a continuous monitoring device can provide the required data. The problem of interpreting the data remains even if a continuous monitor is used, however.

A statistical test that is particularly sensitive in detecting long-term trends, whether these trends are periodic or not, and excessively rapid oscillation in an observed series is the mean square successive difference (MSSD). Briefly stated, it is a test of the null hypothesis that the variance, S^2 , of a series of observations, X , i.e.,

$$S^2 = \frac{1}{n-1} \sum_{i=1}^n (X_i - \bar{X})^2,$$

belongs to the same population as the mean square successive difference of the series, i.e.,

$$\text{MSSD}^2 = \sigma^2 = \frac{1}{n-1} \sum_{i=1}^{n-1} (X_{i+1} - X_i)^2,$$

$\sigma^2/2$ is an unbiased estimate of the variance and is less biased by trends than the usual estimate, S^2 . A disparity between these estimates indicates trends or short period oscillations. In the first case, the ratio σ^2/S^2 will be small since the value of σ^2 will not be increased by the trend as much as S^2 . In the second case (rapid oscillation) both σ^2 and S^2 will increase, but the increase in σ^2 will be proportionately greater.

A basic question in the design of any monitoring system concerns the

limit of required detectability.

Historically, radiation protection requirements have forced the design of highly sensitive monitoring instruments and analytical procedures. In a sense, the same situation continues, but monitoring technology has developed to the point that the designer of a plant now has a sophisticated array of equipment and procedures from which to select.

A system at the least sensitive enough to measure reliably ($\neq 10\%$ at the 95% confidence level) and quickly (< 2 hours) at the safety limit level is required. In some cases an operating guide that is one or two orders of magnitude less than the ICRP value may be used for administrative reasons. At Shippingport, for example, a release limit of 1/10 the ICRP suggested MPC was taken as a design criterion.

The principal factor limiting the detectability of an instrument is the signal to noise ratio with the natural background plus electronic noise constituting the "noise". A lower limit in sensitivity, if required, can be obtained by minimizing the background counting rate through the use of shielding and anticoincidence counting.

The statistics of counting and particularly the correction for background radioactivity are well described in current literature. One point worth emphasizing, however, is that the standard error in a net sample count is the error of the difference between the sample and the background counts. That is, if $A \pm a$ and $B \pm b$ are the reported sample count and the background count, respectively, with their respective errors then,

$$[A \pm a] - [B \pm b] = A - B \pm \sqrt{a^2 + b^2}.$$

As the difference between A and B approaches zero, the relative importance of the error term increases. The count rate error can be expressed as $\pm \sqrt{A/n}$, where A is the total count and n is the counting rate error. The decrease in error obtained by increased counting time has the undesirable concomitant effect of slowing the response time of the monitoring method. Thus, an instrument measuring a concentration of 1×10^{-8} μCi with, say, an accuracy of $\pm 10\%$ at the 95% confidence level for a counting period of 24 hr, requires concentrations of approximately 10^{-3} $\mu\text{Ci/cc}$ for the same accuracy over a counting period of a few seconds.

As stated, the lower end of the range of a monitoring system or the "limit of detectability" is an important factor in the system design. Obviously, there also must be an upper limit to the range as it results from circuit limitations, detector saturation, and coincidence loss. The upper limit is particularly significant in the event of grossly abnormal conditions. The monitoring required during such abnormal conditions is a part of the specialized subject of reactor hazards analysis and excluded from the scope of this document, but it is worth noting that "normal" conditions should be defined so that an alarm can be incorporated into the monitoring system to warn of "abnormal" conditions. If the level of activity, being monitored, continues to rise, a signal from the monitor can be made to activate an automatic protection device. The specific nature of the action taken and

the activity level at which it must be taken require an evaluation of the characteristics of a particular reactor. Again, such evaluations are in the province of hazards analysis, but identification of the range between subnormal and above normal conditions, which monitoring systems are required to span, is an important step in the design of a monitoring system.

Monitoring of Liquid Effluents

The designer's first step in the design of liquid waste monitors is to identify the liquid waste arisings and estimate their quantity and activity. Tables I - IV illustrate the quantities and concentrations of waste arising at four operating power stations. The large volumes of water used by power plants for condensing the used steam (~ 0.3 to 1.5 gal/min/kW) provides a convenient diluent for discharged liquid wastes. From Tables I - IV, it can be seen that the total activity of all liquid wastes from operating plants is extremely small (on the order of 10^{-2} - 10^{-1} Ci/yr). If this amount of activity is diluted with the annual condenser discharge volume ($> 10^{10}$ gal/yr), the average concentration of activity is very low indeed (Table V).

The very low concentrations of radionuclides in the waste streams that have been diluted with the main condenser effluent are impractical to measure. If the wastes are to be monitored, it will have to be done before the wastes are diluted. The waste systems normally employ tanks for holding all possible contaminated wastes temporarily. When one of these tanks is full, it is sampled and the

TABLE I- Shippingport Station (a)

^a Burning rate is 40 pounds per hour.

(a) Reprinted with permission of the Edison Electric Institute. Taken from "Radioactive Wastes Handling In The Nuclear Power Industry."

SOURCE
(Maximum design estimates)

a This tank receives water bleed from the primary system.
b For the design basis, refueling shield water is not considered a waste.
c Laundry will not be installed initially, but system is designed to handle laundry wastes.
d Burning rate is 40 lbs/hr.

TABLE III.
Indian Point Station Waste Plant

TABLE IV. Dresden Station Waste Plant

^a This activity, measured at discharge, is primarily from short-lived xenon and krypton isotopes with half-lives from ten seconds to 12 days. The only significant long-lived isotope is krypton-85, of which about 300 curies per year will be discharged on the maximum basis.

^b This activity is primarily from argon-41.

^c At maximum activity release rates it is estimated that dose rate to off-site personnel will not exceed maximum permissible rates of 0.5 rem per year.

^d Most of the activity is from leakage during scrams -- about 1000 gallons per year, containing one μ c/cc. Daily leakage is less than 10^{-5} μ c/cc.

^e Initially there will be no off-site disposal of solid wastes.

TABLE V. Gross Beta - Gamma Activity of the Liquid Wastes Discharged from Several Operating Power Reactor Plants

	Average Concentration Before Dilution $\mu\text{Ci}/\text{cm}^3$	Average Concentration After Dilution with Main Condenser Coolant $\mu\text{Ci}/\text{cm}^3$	MPC $\mu\text{Ci}/\text{cm}^3$	%MPC
<u>Shippingport</u> ^(a)				
1959				
1960		4×10^{-9}	6×10^{-7}	0.6
1961				
1962				
1963				
<u>Yankee</u> ^(b)				
1961	4×10^{-6}	8×10^{-10}	6×10^{-7}	0.8
1962	2×10^{-6}	5×10^{-10}		0.5
1963	1×10^{-6}	3×10^{-10}		0.3
<u>Dresden</u>				
1961				
1962			6×10^{-7}	
1963				
<u>Humboldt</u>				
8/1/63 to 10/1/63		4×10^{-9}	(c)	
<u>Big Rock</u>				
1/1/63 to 10/4/63		2×10^{-8}	6×10^{-7}	
<u>Indian Point</u>				
1963	$\sim 10^{-5} - 10^{-6}$	$10^{-10} - 10^{-11}$	6×10^{-7}	

(a) From US PHS. Radiological Health Data monthly reports

(b) From "Radioactive Waste Disposal at a Nuclear Power Plant," John Bernsee. May 25, 1964.

(c) The state of California has established an average annual discharge limit of $10^{-8} \mu\text{Ci}/\text{cm}^3$ based on a conservative interpretation of reconcentration factor for edible shellfish

concentration of gross beta-gamma activity determined. It is then possible to determine whether or not and at what rate the waste can be discharged although the rate of discharge is generally not important at the activity levels encountered. In

this case the sampling performs both a process and effluent monitoring function.

The minimum regulatory requirements of liquid effluent monitoring are set by paragraph 20.106 of 10 CFR Part 20 (See Appendix A) which states that no

licensee shall release a concentration of radioactive (licensed) material into water in any unrestricted area in excess of the limits specified in Appendix B, Table B-II. It further states that the concentrations may be averaged over periods not greater than one year. Exceptions to these requirements are possible under paragraph 20.302 of 10 CFR Part 20, but to date no exceptions are in effect at any of the power reactor plants.

The effluent monitoring requirements as set forth in 10 CFR Part 20 and the liquid effluent monitoring required for strictly radiation protection reasons are identical except for two possible exceptions. A periodic radionuclide analysis of the effluent is useful if the dose to persons in the environs is being estimated. Also, if certain radionuclides concentrating in food chains leading to man are present in the waste stream, it may be necessary to measure their concentration. The particular radionuclides that concentrate in the environment are listed, and their movements in the environment are described in the next chapter.

As noted previously, there is normally no reason to routinely identify the individual radionuclides present in a liquid waste because the curie quantities involved are so small that the "unidentified mixture" requirement of 10 CFR Part 20 applies. According to 10 CFR Part 20, if it is known that Sr^{90} , I^{129} , Pb^{210} , Po^{210} , At^{211} , Ra^{223} , Ra^{224} , Ra^{226} , Ac^{227} , Ra^{228} , Th^{230} , Pa^{231} , Th^{232} , and natural thorium are not present, then the MPC is 3×10^{-6}

$\mu\text{Ci/ml}$. If it is known that Sr^{90} , I^{129} , Pb^{210} , Po^{210} , Ra^{223} , Ra^{226} , Ra^{228} , Pa^{231} , and natural thorium are not present, then the MPC is $2 \times 10^{-6} \mu\text{Ci/ml}$. If it is known that Sr^{90} , Pb^{210} , Ra^{226} and Ra^{228} are not present, then the MPC is $6 \times 10^{-7} \mu\text{Ci/ml}$. If it is known that Ra^{226} , Ra^{228} and I^{129} are not present, then the MPC is $1 \times 10^{-7} \mu\text{Ci/ml}$. In the case of power reactor wastes, the absence of Ra^{226} , Ra^{228} and I^{129} can be validly assumed, but the absence of Sr^{90} is not assured so that the "unidentified mixture" limit to be used is $1 \times 10^{-7} \mu\text{Ci/ml}$.

Even though the possible contaminated liquid wastes at all of the power reactors presently operating are collected in tanks, sampled, and released at controlled rates, all of these plants employ a final liquid effluent monitor. Some of the characteristics of these monitors are compared in Table VI.

A recording effluent monitor provides a continuous, automatic, and final record of the quality of an effluent stream. It may also be designed to sound an alarm if the intensity of all radiations or certain selected energies of radiation exceed a certain level. The data produced by a continuous effluent monitor are directly useful in proving compliance with regulatory waste release limits, in public relations matters; and as evidence of safe operation in damage claim suits. It might be argued that the amounts (or concentrations) of radioactive materials actually released during some unusual event (such as an operator discharging the wrong waste tank) must be known so that subsequent

TABLE VI. Liquid Effluent Monitors

Plant	Location	Sensitivity	Detector
Indian Point	Nuclear Boiler	$1 \times 10^{-6} \frac{\mu\text{Ci}}{\text{ml}}$ gamma	2 in. Crystal
	Secondary scale blowdown	activity based on Na^{24}	
	Chemical systems building drain sump	$1 \times 10^{-6} \frac{\mu\text{Ci}}{\text{ml}}$ gamma activity based on Na^{24}	2 in. Crystal
Pathfinder	Effluent	$1 \times 10^{-7} \mu\text{Ci/cc}$ gamma activity based on Cs^{137}	NaI(Tl) Crystal 100 liter chamber
Dresden			
Yankee	Boiler Blowdown		GM tube in 1 ft^3 Detector operates at 250°F
Elk River	Effluent	$5 \times 10^{-6} \mu\text{Ci/cc}$ based on Zn^{65} 0.375 MeV β 55% 1.12 MeV γ 45%	Crystal
La Crosse	Effluent	$5 \times 10^{-6} \mu\text{Ci/cc}$ based on Zn^{65} 0.375 MeV β 55% 1.12 MeV γ 45%	Crystal
Piqua	Effluent	$5 \times 10^{-6} \mu\text{Ci/cc}$ based on Zn^{65} 0.375 MeV β 55% 1.12 MeV γ 45%	
Humboldt Bay	Waste discharge line to circulating water discharge canal	Nominal sensitivity $5 \times 10^{-6} \mu\text{Ci/ml}$ (a)	NaI(Tl) crystal is shielded in-line sample chamber

(a) In practice, actual sensitivity depends on the degree of contamination of the sample chamber. The chamber is flushed and/or removed for cleaning when sensitivity deteriorates significantly.

releases can be adjusted to not exceed the annual average within permissible limits. The same information could be obtained by collecting and analyzing samples after an alarm had indicated an unusual event. Such samples prob-

ably would be collected anyway for individual radionuclide analysis.

The importance of continuous effluent monitors would increase considerably if the liquid wastes were released without first temporarily

retaining the wastes. In such a system a continuous effluent monitor would provide the only information about the instantaneous condition of a waste stream. If information is to be obtained by analyzing samples of the waste stream in a laboratory, some form of standby waste retention or recirculation capability would probably have to be provided. This would delay the decision of abnormal waste disposal until the sampling results are available.

Gaseous Effluent Monitoring

A reactor plant is prepared to control the release of the radioactive gases and particles conceivable from an accident. The control procedures and equipment, required during these hypothesized situations, are considerably more than adequate to control the normal release of gaseous wastes. Thus, the principal purpose in monitoring normal gaseous effluent releases is to demonstrate compliance with federal regulations.

These minimum requirements as set forth in 10 CFR Part 20 frequently are augmented by requirements imposed as a condition of operating license approval. The additional requirements are initially proposed by the license applicant and, if deemed adequate by the regulatory agency, are made a condition of compliance. Safety limits are usually the important part of the additional requirements. Sometimes, monitoring procedures and equipment also may be specified, particularly if radioiodine is to be released.

The calculation of maximum permissible stack release limits involves a number of complex variables and re-

quires reliable meteorological data. The calculations of chronic or normal release limits are seldom if ever made for that purpose only but, as mentioned earlier, are an outgrowth of the extensive effort required to estimate the consequences of emergency situations.

The gaseous waste release limits currently in effect at power reactor stations are shown in Table VII along with other data pertinent to the monitoring equipment and procedures used at these plants.

The form and extent of evidence required to prove compliance with regulatory requirements has not been established; however, as a minimum it must be at least sufficient in nature and quantity to convince the Division of Compliance inspectors that no regulatory requirements have been violated. The most convincing evidence would be a record of the quantity and concentration of the radionuclides released. Although, as noted previously, only proof that safety limits have not been exceeded should be needed to prove compliance.

ENVIRONMENTAL MONITORING

Objectives

The second report of the Federal Radiation Council contains a noteworthy statement relating to the objectives of environmental monitoring:

"The objective of the control of population exposure from radionuclides occurring in the environment is to assure that appropriate RPG's are not exceeded. This control is accomplished in general either by restrictions on the entry of radioactive materials into the environment or through measures designed to limit the intake of

TABLE VII. Characteristics of Gaseous Effluent Monitors at Several Power Reactor Plants

Reactor	Height, ft Bldg. Stack	Dilution Factor Licensed	Average Discharge Concentration	Number of Gas Monitors	Sample Volume	Types of Monitors
Yankee	149	None	4.4×10^{-8} $\mu\text{Ci/cc}$	3	-	β Crystal β γ GM
Indian Point	400	1000 (gas) 100 (particulate)	$\mu\text{Ci/sec}$			
Dresden	130	300,000 $\mu\text{Ci/sec}$	4.77×10^{-5}	4	0.68 cc 0.1 ft ³	γ Crystal (b) γ Crystal γ Crystal β Crystal
Elk River	~100	25,700 $\mu\text{Ci/sec}$	-			
Big Rock						
Pathfinder	107	Not specifically licensed	-	2 off-gas 2 stack gas (2)	10 x 12 in. diameter	Thin anthracene Crystal Anthracene scintillation ion chamber
Humboldt Bay	250		-			

TABLE VII. (contd)

Reactor	Sensitivity for 2 times Background, $\mu\text{Ci/cc}$	Calibration Sources	Frequency of Calibration	Radiation Measured and mg/cm^2	Scale Range	Curies Released in 12 Months	Stack Flow, cfm	Licensed Release Rate $\mu\text{Ci/sec}$
Shippingport	2×10^{-7} (a)	Kr ⁸⁵ Co ⁶⁰	(P) Monthly	Beta 2 Gamma 30	5 decades 3 decades	0.00154	30,000 cfm	1000 x MPC
Yankee	4×10^{-6}							
Indian Point		Cs ¹³⁷ N ¹³	(P)	Gamma		4,200	45,000	700,000
Dresden		Co ⁶⁰ Th ²⁰⁴	Yearly	Gamma Beta	logarith. 5 decades	3 to 5 curies/yr	3,200	
Elk River								10^6 $\mu\text{Ci/sec}$
Big Rock								
Pathfinder		Kr ⁸⁵	Monthly	Beta	4 decades	-	25,000 (min)	[83,500 $\mu\text{Ci/sec}$ Noble gas
Humboldt Bay		Actual off-gas	Daily grab samples	Gamma Gamma	(1)	(3)	12,000	50,000 Noble gas 0.18 halogen and particulate

(a) Due to background increase, this value is now nearer $\sim 10^{-6}$.

(b) In addition to a gas and particulate monitor, Dresden recently installed an ion chamber monitor on the off-gas line.

(1) 5 decade (10^{-4} to 10 curies/sec) on both off-gas and stack gas monitoring systems

(2) In addition to these noble gas monitoring systems, a halogen and particulate sampling system is provided to obtain samples for routine laboratory analysis.

(3) A comparison of the curie releases between BWR and PWR plants is not valid because of the large difference in half-life of the noble gas mixtures from the two types of plants. The important consideration is the instantaneous number of curies in the atmosphere which determines dose. During the last year of operation at Humboldt Bay, the average stack release rate was 6200 $\mu\text{Ci/sec}$ which is 12.4% of the 50,000 $\mu\text{Ci/sec}$ release rate limit.

such materials by members of the population. The most direct means of evaluating the effectiveness of control measures is the determination of the amount of radioactive material in the bodies of the members of exposed population groups. Although the determination of such body burdens may at times be indicated, in routine practice potential exposures will generally be assessed on the basis of either one or a combination of two general approaches: (1) calculations based upon known amounts of radioactive material released to the environment, and assumptions as to the fraction of this material reaching exposed population groups, or (2) environmental media."

Of particular importance at this point is the statement that environmental media can be monitored to determine the effectiveness of waste control procedures. This is the primary reason for an environmental monitoring program at a power reactor plant. There are also less important for monitoring reasons such as the public relations demonstration that concentrations of radioactive materials released by a nuclear power plant are so small that they are impossible to measure, but these reasons are virtually the same as the secondary reasons for effluent monitoring as discussed in the previous chapter and will not be repeated here.

The point was also made in the previous chapter that the closer the actual quantities of radioactive materials released from a plant approached the operating limits, the more precise must be the effluent monitoring procedures employed. The same rationale carries over into the environmental monitoring used for assessing the effectiveness of waste control proce-

dures: The closer the quantities (curies) of waste actually released approach release limits, the more important it becomes to measure the dose delivered to persons in the environs.

Requirements

The movement of radionuclides from a point of release to a point of exposure can be as simple as the diffusion of a cloud of noble gases or it can be a complex pathway through several trophic levels. Figure 1 illustrates the pertinent factors. Parker et al hypothesized a variety of possible

FIGURE 1. Schema of Pertinent Factors Involved in the Exposure (Internal) of Persons from the Release of Radioactive Wastes

sources of exposure in the environs of an atomic energy installation (Figures 2-4). The designer of every new facility should consult this list to determine if any of the potential

sources of exposure will have to be evaluated for the facility he is designing. Most of the pathways listed are obviously irrelevant to power reactor practice. A few of the pathways

FIGURE 2. Exposure Pathways for Radioactive Wastes in Surface Waterways

FIGURE 3. Exposure Pathways for Radioactive Wastes in the Atmosphere

FIGURE 4. Exposure Pathways for Radioactive Wastes in the Ground

involving the biological concentration of radionuclides may need to be studied before they can be dismissed as inconsequential. Depending on the particular conditions at an installation, it may be necessary to maintain surveillance over one or two pathways during operation of the plant in order to estimate the radiation exposures being received by persons in the environs via these pathways. The surveillance requirements of the terrestrial and the aquatic environments differ somewhat, and should be considered separately.

Certain radionuclides are known to be capable of concentrating biologically to significant levels in the terrestrial environment; they are: strontium-89 and 90, barium-140 - lanthanum-140, cesium-137, iodine-131, zinc-65, radium-226, iron-55, and phosphorous-32. The last three of the nuclides are not produced in significant amounts in current power reactor practice, so possible exposure to them via terrestrial pathways is relatively unimportant.

The principal pathway of exposure for the first eight radionuclides listed in the previous paragraph is through milk to man and can be diagrammed as follows:

Under normal reactor operating conditions, the direct exposure of people from these radionuclides and the effect of contaminated irrigation water can

be safely ignored. The particular characteristics of radioiodine makes its release to the environment, via the atmosphere, of much more importance than the release of other radionuclides contained in gaseous reactor effluents. For example Russell states that experience from weapons tests and the Windscale accident show that, in atmospheric contamination with fresh fission products, the dose to the thyroid from iodine-131 and iodine-133 exceeds by considerably more than an order of magnitude that delivered by absorbed nuclides to other tissues.

Zinc-65 is formed by neutron activation and does appear in the liquid wastes of some power reactors e.g., Humboldt Bay. It is an isotope of an element utilized in the metabolism of most aquatic life forms. Its half life is long enough for it to enter complex transport and transfer processes in various food chains. Shellfish, particularly oysters, concentrate zinc avidly. Consequently, this exposure pathway can become important in situations where activated corrosion products are released into the ocean or into tidal bays. The movement of zinc-65 released to the Columbia River at Hanford has been studied rather intensively and well reported.

The factors involved in the transfer of iodine from the atmosphere into the food chain have been given a great deal of attention. Empirical relationships for the important factors appear in Table IV. The data appears as collected and reported by Cowser, with the exception that the column "milk to air" concentration ratio has been added by

manipulating concentration ratios thusly:

$$\begin{aligned} \text{Milk to air } & \frac{\text{pCi/liter}}{\text{pCi/meter}^3} \\ &= \text{Milk to grass (pCi/liter/pCi/kg X)} \\ & \quad \text{X Grass to air (pCi/kg/pCi/meter}^3\text{)} \end{aligned}$$

The data presented in Table VIII would seem to support the use of a conservative milk to air concentration ratio of 700, although the values are highly variable. Thus, the above concentration factor is useful in establishing conservative effluent rates, but cannot be used effectively in predicting concentrations of iodine in milk by using air monitoring data. At locations where radioiodine is released into an agricultural community containing dairy cows, the milk from these cows should be monitored for radioiodine. Before discussing the specific requirements of monitoring milk for radioiodine, perhaps the fate and relative importance of in the terrestrial environment of the other radionuclides (listed above) should first be considered.

As in the case of iodine release, cow's milk is the principal terrestrial exposure pathway for strontium and cesium. It must be noted that there could be locations where the grass-animal-meat-man pathway is nearly as important for the movement of cesium as the milk pathway. Milk can be expected to contain approximately 5% of the iodine-131 and 0.5% of the strontium in the diet of a cow. While cesium does appear in milk, it does not tend to concentrate to the same degree as iodine, nor remain in the body of the milk drinker as long as strontium.

Therefore, it has less radiation protection interest than do iodine and strontium. Cesium reacts with soil and is available to a plant only through foliar deposition. Iodine and strontium, however, can enter herbage through the soil and root matter, as well as through foliar absorption.

Barium-140 - lanthanum-140 also appear in the milk pathway, but their radiological significance is considerably less than it is for iodine because of the manner in which they are metabolized.

From the foregoing three paragraphs, it can be concluded that milk is the media to be sampled if the degree of reconcentration of radionuclides in the terrestrial environment is to be assessed. If a reactor plant is located in an area where no milk is produced, some consideration should be given to other possible terrestrial exposure pathways, but it is doubtful that any of them will be significant. Indeed, the milk pathway is significant only if fairly large amounts of iodine and/or the other radionuclides (listed above) are released. (Release of 10 mCi/week of I^{131} produces concentration in milk of ~ 2 pCi/l.) If the collection and analysis of milk samples is indicated at a particular site, the sampling location and the size and frequency of the samples must be established.

The first step in determining whether or not a milk sampling program should be instituted and the first step in actually starting such a program is to identify the loca-

TABLE VIII. I^{131} Concentration Ratios Between Various Media

Location	Date	Source of I^{131}	Ratios of I^{131} Concentrations		
			Grass to Air ($\mu\text{Ci/kg}$ per $\mu\text{Ci/m}^3$)	Milk to Grass ($\mu\text{Ci/l}$ per $\mu\text{Ci/kg}$)	Milk to Air ($\mu\text{Ci/l}$ per $\mu\text{Ci/m}^3$)
SRP ^(a)	1959	Weapons fallout, dry grass	20,000		
	June 1961	SRP release, dry dry grass	37,600	0.018	676
	Sept. 1961- Jan. 1962	Weapons fallout, dry grass	19,500	0.021	409
HAPO ^(b)	April - Oct. of 1961 and 1962	Weapons fallout, fresh grass			
		Weapons fallout, dry grass	21,000	0.03	630
BNL ^(c)	Sept. - Dec. of 1962	Weapons fallout, fresh grass		0.07	
Rome, Italy ^(d)	Nov. 1961	Weapons fallout, fresh grass		0.021	
Windscale, England ^(e)	Oct. 1957	Windscale release, fresh grass		0.09	

- (a) W. L. Marter. "Radioiodine Release Incident at the Savannah River Plant," Health Phys., vol. 9, pp. 1105-1109. December, 1963.
- (b) J. K. Soldat. "The Relationship Between I^{131} Concentrations in Various Environmental Samples," Health Phys., vol. 9, pp. 1167-1171. December, 1963.
- (c) A. P. Hull. "Vegetation Retention and Vegetation - Milk Ratios of Fallout I^{131} ," Health Phys., vol. 9, pp. 1173-1177. December, 1963.
- (d) A. A. Cigna, M. M. Ferraris, F. G. Giorcelli, and C. L. Papucci. "Composition and Deposition in Italy of Fallout from USSR Explosions, Fall 1961," Health Phys., vol. 9, pp. 319-331. March, 1963.
- (e) H. J. Dunster, H. Howells, and W. L. Templeton. "District Surveys Following the Windscale Incident, October 1957, Proceedings of Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958, vol. 18, p. 296. United Nations, New York, 1958

tions of milk producing cows that possibly can be affected by material released from a nuclear plant. Commonly, milk cows are either part of a large dairy herd or are kept by families to provide milk products for one or at most a few families. The milk produced by many herds in a milk shed are combined for processing and distributing to population centers.

The radiation dose received by small groups of individuals living in the environs and obtaining milk products from one or a few cows has prime radiation protection interest. The doses received by persons who drink milk collected from an entire milk shed must be considered of course, but the possibility of normal reactor operations causing more than an infin-

itesimal dose via this route is highly unlikely.

If a milk sampling program is indicated and appropriate sampling stations (dairy cows) have been located, the next step is to establish a sampling frequency. Because the concentration of radionuclides in the milk can be expected to vary from time to time due to variations in the amount of the radionuclides released, diet of the cows, etc., it is important that the sampling period be equal to or shorter than the periods of variation in the milk to be sampled. Since iodine-131 is the shortest lived radionuclide of interest in milk sampling and has a half life of ~ 5 to 6 days in the exposure pathway, the sampling period should be short enough to assure that individual variations of that nuclide are properly represented in the annual average. A one-week sampling period would definitely include all variations, but the error involved in sampling on a two-week or twice monthly period should not be important, particularly after data from many months are available. After several years of data collection a pattern of variation should become apparent, and the sampling frequency can be altered accordingly.

The precision required in a milk sampling program is dependent upon the error that can be tolerated, and it increases as the concentration of the radionuclide of interest approaches some maximum permissible level. For example, during normal operations the effect of the radioiodine released by a reactor plant probably will not be detectable in milk even during periods

of little or no fresh weapons fallout. The normal concentration of iodine-131 in milk for most of the United States at such time can be expected to be approximately 10 pCi/l or less. A daily intake of milk containing approximately 80 pCi/l results in a dose of 0.5 rem/yr to a 2 g thyroid of a 2 yr old child. If the monitoring data are used to calculate the radiation dose received by persons who drink the milk, it is desirable to know with considerable certainty (degree of confidence) that a maximum permissible dose is not exceeded. Stated more specifically, during normal conditions a standard error of $\pm 100\%$ can be tolerated. Such an allowable error enables a very high degree of confidence to be obtained without exceptional effort.

The minimum size of a milk sample that must be collected is set by the requirements of the analytical technique to be used. Iodine-131 is most conveniently measured by counting its 0.36 MeV gamma photon. The milk can be analyzed directly in a large NaI(Tl) crystal. A detection limit of 50 pCi/l for a 500 ml sample is obtained at Battelle Northwest Laboratories by this means. The detection limit is lowered to 2 pCi/l by passing several gallons of milk through a large bed of anion-exchange resin and measuring the activity accumulated on the resins. Chemical and radiochemical recoveries for the separation average 90%, and precisions are typically $\pm 10\%$ at the 95% confidence level.

In addition to showing short term fluctuations, a milk sampling program can be used to indicate relatively

long term trends such as the build up of strontium-90 in pasture land around a reactor plant. Collecting samples four times a year should be sufficient for this purpose. In view of the extremely minor amounts of strontium-90 released from reactor plants, the probability of a significant build up is highly unlikely, and information coming from a milk sampling program will be largely negative, i.e., no effluent strontium is observed. The problem is to differentiate between effluent strontium and fallout strontium.) In a sense then, the radiation protection value of such sampling is very slight and if there is no reason for doing the work, e.g., public relations, the work probably need not be done by the reactor plant operator. However, public health agencies may be interested in such low level counting as a part of their responsibility to maintain surveillance over environmental contamination from various sources.

It has become somewhat traditional for atomic energy establishments to collect samples of media, other than milk, that are involved in terrestrial exposure pathways such as, vegetation and soil. Some worthwhile purpose may be served by this practice where relatively large amounts of radioactive materials are released. However, the amounts of radioactive materials released from power reactors is so small and indistinguishable from background that the dose contribution from the reactor operation cannot be calculated by using the data from analyzing soil, vegetables, etc. It is valid to state, therefore, that sampling such media

clearly does not have a radiation protection purpose. The purpose is probably best described as the development and improvement of radiation protection techniques and practices. Presumably, a power reactor plant management has little interest in environmental monitoring research and development, and the sampling of terrestrial media other than milk need not be considered (at least for radiation protection purposes). There may be valid public relations, public health and liability reasons for such sampling, however.

The fate of radionuclides released into an aquatic environment must be considered in the siting, design, and operation of every power reactor. Because current power reactor technology requires large quantities of cooling water, the reactor plants are located near sizable bodies of water (usually rivers). The amounts and concentrations of radionuclides released into these bodies of water by the reactor plants are extremely small when compared to the practices of major atomic energy establishments which are carried out with proved safety. In this regard, a comparison of the quantities of low-level waste released to rivers and lakes in North America made by Parker et al, points up the virtually negligible quantities of waste released by power reactors. Quantities of radioactive materials released are listed in Table IX, and the estimates of the exposures received by people from low-level wastes discharged into North American rivers are summarized in Table X. The report cited contains several conclusions which, although rather general, are nevertheless

pertinent to this portion of the text and are worth repeating in toto:

"The radioactive materials that have entered the rivers adjacent to the major North American atomic energy sites have received continuous attention so that their fate in the environment and the several important exposure pathways could be evaluated. A gratifying outcome of this is confirmation that the total dose to the public from multiple environmental sources has been well within appropriate limits at all sites. The general tenor of the

environmental studies indicates sound and continuous control of the radioactive effluents from these sites.

In comparison with the quantities of radioactive materials released from the major sites, the releases from the nuclear power stations have been exceedingly small and the dose to the public has been negligible. Methods are thus available which can provide the necessary continued reliable control of liquid effluents associated with the expected growth of the nuclear power industry in North

TABLE IX. Quantities of Low Level Radioactive Wastes Added to Streams, curies per day

		SITE							
Nuclide		Half-Life	Chalk River		Hanford		Oak Ridge	Savannah River	Nuclear Power
Activation Products	Cu ⁶⁴	13 h	-		200 ^(a) - 1000 ^(b)		- ^(d)	-	-
	Na ²⁴	15 h	(Trace)	T-10	200	-1000	-	-	-
	As ⁷⁶	26 h	T- 5		50	- 300	-	-	-
	Np ²³⁹	2.3 d	-		200	-1000	-	2 ^(c)	-
	P ³²	14 d	0.01 - 0.1		20	- 70	-	-	-
	Cr ⁵¹	28 d	-		600	-2000	-	3.5	T
	Fe ⁵⁹	45 d	-		T		-	-	T
	Co ⁵⁸	71 d	-		T		-	-	T
	S ³⁵	87 d	0.001		T		-	0.4	-
	Zn ⁶⁵	250 d	0.002		30 - 100		-	0.09	-
Co ⁸⁰		5.3 y	T- 0.05		1 - 2	0.04 - 0.2	0.01	T	
H ³		12 y	0.01 - 20		T	-	205	-	
Fission Products	I ¹³¹	8 d	-		1 - 3	0.001-0.01	0.1	T	
	Ba ¹⁴⁰	13 d	-		T	-	0.1	T	
	Nb ⁹⁵	35 d	-		-	0.002-0.2	-	-	
	Sr ⁸⁹	50 d	-		T	-	0.9	-	
	Zr ⁹⁵	65 d	-		T	0.001-0.1	0.1	-	
	Ce ¹⁴⁴	285 d	0.005-0.015		-	0.003-0.1	0.1	-	
	Ru ¹⁰⁸	1 y	0.002		-	1-5	0.03	-	
	Sr ⁹⁰	28 y	0.001-0.005		0.1	0.02 -0.2	0.03	- ^(e)	
	Cs ¹³⁷	30 y	0.002-0.02		T	0.01 -0.2	0.3	-	
	Total Beta (Exclusive of H ³)		0.05 -15		2000	1 -6	(0.5 in river)	10 ⁻⁵ - 0.01	
Receiving Stream		Ottawa River		Columbia River	Clinch River	Savannah River	Various		
Flow Range		3-13		10-75	0.6-2	1.4-8	1.10		
10 ¹⁰ l/d Average		6		27	1	2.5			
Measurement Point		Ottawa River Process Sewer Perch Lake		Columbia River (Pasco)	White Oak Creek	Storage-Basin discharge	Various waste streams		

(a-b) Where a substantial variation is reported, both the low (a) and high (b) values are listed.

(c) Where a yearly average is reported, or there is little variation, only one value is listed.

(d) (-) indicates the nuclide is not reported. It may be present but in amounts that are trivial in relation to other nuclides encountered.

(e) Fallout contributed from 0.1 to 1 curie of Sr⁹⁰/day to large rivers of North America in 1963.

TABLE X. Significance of Exposure from Various Sources

Site	Nuclides of Greatest Interest	Mode of Exposure	Critical Organ	Type of Person Receiving Greatest Exposure	Percent of Limit (a)	Year	Reference Limit
Chalk River	Sr ⁹⁰	Drinking Water	Bone	Pembroke-Resident	<1 ^(b)		ICRP Population at large
	P ³²	Fish	Bone	Fisherman	<0.1	1963	ICRP Group B, c ^(c)
Hanford	P ³²	Fish and Irrigated Crops	Bone	Fisherman-Farmer	<40		ICRP Group B, c ICRP Population at large
	As ⁷⁶ + Np ²³⁹	Drinking Water	GI Tract	Pasco-Resident	<8	1963	
	I ¹³¹	Drinking Water	Thyroid	Pasco-Child	<6		FRG ^(d) Exposed Population
Oak Ridge	Sr ⁹⁰	Fish	Bone	Fisherman	<30 ^(f)		ICRP Group B, c
	Sr ⁹⁰	Drinking Water	Bone	Clinch River - Resident ^(e)	<5	1961	ICRP Group B, c
	Ru ¹⁰⁶	Drinking Water	GI Tract	Clinch River - Resident ^(e)	<5		ICRP Group B, c
Savannah River	H ³	Drinking Water	Whole Body	Savannah River - Resident ^(e)	<4		ICRP Genetic Apportionment
	I ¹³¹	Drinking Water	Thyroid	Savannah River - Resident ^(e)	<0.3	1963	ICRP Population at large
	Sr ⁹⁰	Fish + Water	Bone	Fisherman	<1 ^(b)		ICRP Population at large
Power Station	Co ⁶⁰	Fish	GI Tract	Fisherman	<0.01		ICRP Group B, c

(a) Excludes natural background, medical, and atmospheric pathways, but includes contributions to the same organ from other water-borne sources.

(b) Most of the Sr⁹⁰ contributing to this exposure was from fallout and not plant operations.

(c) Recommendation adopted September 9, 1958. Group B, c is "members of the public living in the neighborhood of controlled areas".

(d) Federal Radiation Council (U.S.) Recommendations of September 1961.

(e) A hypothetical person who drinks untreated river water. No such person has been found.

(f) Assumes that whole fish, including bones is eaten. If only the flesh is eaten the estimate is <6%.

America and such effluents need not be a deterrent to that growth."

Reactor plants normally restrict the concentration of radionuclides in liquid effluent streams to a level less than the pertinent maximum permissible concentrations (MPC) suggested by the ICRP and other advisory organizations. The actual exposure received by persons in the environs resulting from the release of wastes has considerable radiation protection value and should be estimated. The use of the MPC values as upper limits does provide rather reliable assurance that the doses received by persons exposed to the effluent, directly or after it is diluted in the environment, will not exceed a maximum permissible value.

Some of the released radionuclides may be reconcentrated in the environment, and the possible exposure to these in concentrated form requires attention.

The radionuclides reportedly capable of concentrating in aquatic environments are: sodium-24, copper-64, iron-59, phosphorous-32, strontium-90 - yttrium-90, zinc-65, cobalt-57, cobalt-60, iron-55, and manganese-54. The reported concentration factors range as high as four orders of magnitude for phosphorous-32 in the Columbia River. While the reported cases are unusual and do not represent circumstances at power reactors, a sustained or repeated release of any of the radionuclides listed above should be accompanied by an investi-

gation of the resulting exposure potential.

Knowledge of local dietary habits is vital to an estimate of the relative hazard of a given level of contamination in any food product, such as fish. The practice of the Hanford Plant provides a good example of how such dietary information can be obtained and used; however, the amounts of radioactive materials released by power reactor plants are many thousands of times less than those released to the Columbia River. At Hanford, conducted creel surveys have revealed that a few avid fishermen claim to eat as many as 200 fish meals per year of fresh Columbia River fish. The relative amounts of the several species of fish eaten were also determined by the surveys. To estimate the exposure to the fish eaters, it is assumed that a fish meal consists of 200 g of fish flesh, an annual consumption of 40 kg/yr; and the consumption is prorated among the various species. The average concentrations of radionuclides (phosphorous-32 is the only significant radionuclide) is determined by catching and analyzing samples of approximately 100 fish/month. Using these data and the stated assumptions, the dose received by fish eaters is estimated.

Excluding very unusual circumstances, the only credible aquatic exposure pathway leads through fish to man, although the fish may be shellfish. The important monitoring data required from such a pathway, in addition to the dietary data mentioned in the previous paragraph, are the concen-

trations of radionuclides in fish that are eaten. It seems highly unlikely that a significant exposure will occur via fish and as a result of the normal operation of a power reactor. However, if such a situation did occur, the frequency with which fish were collected for analysis should be equal to or shorter than any period of fluctuation of the radionuclides concentration in the fish. Monthly samples might be indicated; normally, collecting and analyzing a sample of fish at a time when the concentration of radionuclides should be at a maximum is sufficient to prove that eating fish results in no significant dose. Some monitoring experience may be required to identify the period of maximum concentration in the fish. A combination of low water level and rapid fish growth, such as often occurs in late summer, might produce the maximum radionuclide concentration in the fish.

Again, the size of sample required depends on the limit of detectability of the analytical technique and the concentration of radioactivity in the fish. Actually, only the radioactivity of the portion of the fish normally eaten, viz., the flesh and the bones of such fish as sardines and salmon is really of interest, and presumably, a sample of approximately 100 g would be sufficient. At Hanford, a minimum of 20 pCi of P^{32} /g of fish can be seen by gamma scan, and this limit can be lowered with beta counting.

The precision required in the fish sampling is dependent on the same factors as described for milk sampling.

In view of the considerable error involved in estimating the amount of fish people eat, it is difficult to foresee a lack of precision in the analysis of the fish ever becoming the most important error.

EQUIPMENT AND PROCEDURES

Liquid Wastes

There are two possible approaches to the problem of monitoring liquid effluents. One approach is to place a detection probe in the liquid to be monitored and to make measurements directly, and the second is to remove a representative portion of the medium for subsequent treatment in a laboratory or by a specially designed device such as a continuous monitor.

Because of the difficulty in discriminating between permissible levels of contamination and natural background, the usefulness of in-stream or in-tank monitors is severely limited for the measuring the very low contamination levels normally encountered in power reactor practice. For example, the average energy dissipation (rads/hr) of a concentration of mixed fission products ($\Sigma E = 0.75$ MeV) equal to the 10 CFR Part 20 MPC for an unidentified mixture of radionuclides (10^{-7} μ Ci/ml, assuming uniform distribution and an infinite volume, is: 10^{-7} μ Ci/g $\times$ 3.7×10^{-4} dis/sec/ μ Ci $\times$ 0.75 MeV/dis $\times$ 1.6×10^{-6} erg/MeV $\times$ 3.6×10^3 sec/hr $\times$ 10^{-2} rad/g/erg or 1.6×10^{-7} rad/hr. Since the average cosmic ray background is $\sim 3.10^{-6}$ rad/hr, [Lowder and Solon (1956)] it is obvious that an in-stream direct reading monitor would be useless in such a situation.

As mentioned previously, a prevailing waste management practice at power reactor plants is to collect all possible contaminated liquids in tanks for measurement and subsequent release or treatment. The only sampling involved, therefore, is obtaining representative samples from the tanks.

As experience is gained in operating power reactor plants, the procedures employed, including waste management procedures, will undoubtedly change. The necessity for collecting all liquid wastes in tanks will certainly be challenged. Alternative techniques will require monitoring of flowing streams of possibly contaminated water.

Tank Monitoring

The most obvious and simplest method to sample the contents of a tank is to manually withdraw a dipperful. In reactor plants, the access to collection tanks is usually inconvenient, if not actually impossible. For this reason and because of possible contamination and exposure to the persons who would take the sample, direct sampling is not desirable. A more desirable and more convenient method of sampling is to utilize the pump and piping system required to transfer the contents of the tanks.

Waste collection tanks are invariably located at the lowest levels where they act as sumps and the contents must be lifted to be transferred and discharged. The position head on the discharge side of the pump can be used to withdraw a sample from the system. A piping arrangement for recirculating the tank contents during the sampling period may be necessary. Such an

arrangement also provides an effective method for mixing the tank contents if inhomogeneity of the contents is a factor of any consequence.

The usefulness of continuous monitoring devices, which directly measure the radioactivity of the contents of a tank, are limited by their inability to discriminate between low concentrations of radioactive materials and background radioactivity. Such instruments can detect and measure concentrations of radioactive materials which are about 10^2 times the MPC's, and have rather wide application for detecting, measuring and actuating an alarm if abnormally high levels of radioactivity occur.

Pipeline Monitoring

Most of the samples withdrawn from power reactor plant piping systems are withdrawn for purposes of process control. That aspect of monitoring is specifically excluded from the scope of this document (as stated in the first section). Accordingly, the discussion under this subheading is directed toward the collection of samples of effluent material. It is assumed that such effluents are maintained at pressures at or slightly above atmospheric pressure and at temperatures less than 100 °C. These conditions exclude the sampling of pressurized coolant loops as well as boiler blow-down.

The simplest (and most obvious) means for removing samples from the piping systems is to use valves installed at appropriate locations in the system. Ordinarily, these valves simply control the flow from a hole in the pipe and, therefore, material from

the inner surface of the pipe. If the composition of the material in the pipe varies across the pipe section, such a sample may not be representative. In this case, a representative sample can be obtained by extending a perforated sampling nozzle into the pipe (Figure 5).

FIGURE 5. Sampling Nozzle for Sampling from a Pipe (ASTM Spec. Pub. 148-D)

The inlets of the nozzles must face upstream, and the total cross-sectional areas of the perforations should be such that the flow of effluent in the sampling line is proportional to the flow in the pipe.

The valves can be operated manually. They can also be solenoid valves which, when activated by a timer, open at pre-selected times or frequency. If adequate pressure is not available, a pump may be used to circulate a portion of the stream to the level of the sampling point and collector. An integrated sample can thus be obtained automatically, which will be more representative of the pipe contents than an infrequent grab sample.

One technique for automatic sampling is a jet pump sampler as illustrated in Figure 6.

The jet pump (A) circulates a sample from the effluent pipe continuously and returns it to the pipe. The solenoid valve (B) opens when energized by

FIGURE 6. Jet Pump Sampler

the timer (C), and delivers a small constant sample to the container (D). It is also possible to vary the sample size and frequency in direct relation to the rate of flow by arranging for a flow rate sensing device to actuate the solenoid.

Another automatic sampling device employs an air lift as shown in Figure 7.

A cylinder (A) with a flapper valve is submerged in the line to be sampled.

FIGURE 7. Air Lift Sampler

When the timer opens solenoid valve (C), air pressure in (A) closes the flapper valve and raises the water in (A) and delivers it to the chamber (E). The air pressure also closes the drain in the standpipe (F) through operation of air cylinder and stem (D), permitting the standpipe to fill to overflowing; the excess is sent to waste. After a time delay, the solenoid valve is deenergized, and the system returns to its original state. A turntable (G) is provided to permit segregation of one hour's samples from the next. The turntable is automatically advanced to the next container on signal from the timer. An advantage of this system is the freedom from trouble with the intake cylinder as compared to a pump.

Whenever pipe systems are sampled, accurate flow rate information is important in determining the portion of the total flow represented by the samples. The flow of fluids in a completely filled pipe can be accurately measured by

- Differential head devices such as Venturi tubes, orifices, and nozzles
- Propeller-type meters
- The recently developed magnetic flowmeter.

Continuous Pipeline Monitors

A detector mounted on the outside of a pipeline can provide useful information when used to monitor process streams containing concentrations of gamma emitting nuclides above $\sim 10^{-5}$ $\mu\text{Ci/ml}$. This arrangement is used primarily to detect fuel element failures. The sensitivity does not allow the exact concentrations of radionuclides in most effluent streams

to be measured.

Such a device can indicate whether or not a level of activity in the effluent equal to or greater than its limit of sensitivity has been reached. Consequently, such a device can be used to actuate an alarm should a significantly high release rate occur. If a large dilution capability is available, the ability of such a device to indicate a concentration of gamma emitting radionuclides equal to or exceeding the limit of sensitivity of the instrument can also be used in estimating whether a permissible concentration at the point of discharge has or has not been exceeded. For example, if the main condenser coolant provides a 10^4 dilution factor the permissible concentration in the line discharging to the coolant stream is about 10^{-3} $\mu\text{Ci}/\text{cm}^3$ for an unidentified mixture of radionuclides. Such concentration just might be measurable with an external detector. To use this monitoring approach would require that the gamma activity in the effluent, measured by the external detector, correlate with the total activity in the effluent and some development work would be necessary.

The sensitivity of an external detector arrangement can be improved if the absorbing pipe material is eliminated and the detector is immersed in the medium to be monitored. For reasons explained previously, an immersed detector requires a radionuclide concentration of 10^{-5} - 10^{-6} $\mu\text{Ci}/\text{cm}^3$ to produce a signal equal to background. The average concentrations in undiluted power reactors range from 10^{-6} to 10^{-8} (Table V). Consequently,

the usefulness of immersed detectors in effluent monitoring is limited to actuating alarm circuits if the level of activity rises, but they could also be used either to indicate that permissible levels had not been reached or if they had, to measure the level as discussed in the previous paragraph.

A liquid effluent gamma monitor which utilizes an immersed scintillation detector was developed for use at the Plutonium Recycle Test Reactor at Hanford. The instrument, uses solid state circuitry, a four-decade logarithmic response count rate meter, and additional direct metering circuit. The system was developed to monitor a range from approximately 10^{-4} gamma $\mu\text{Ci}/\text{ml}$ to 5 gamma $\mu\text{Ci}/\text{ml}$. Photographs of the detector probe, readout meters and trip selectors are shown in Figures 8 and 9. The detector scintillator is a 2 in. diam by 2 in. long cylinder of terphenyl in polyvinyltoluene. The multiplier phototube anode is capacitively coupled to the pulse amplifying and counting instrumentation, and is also directly coupled to additional circuitry that provides linear high radiation level indication and an electronic alarm trip. This dual method provides a sensitive pulse counting circuit combined with a direct coupled linear circuit to prevent possible AC blocking at high radiation exposure.

An ideal immersed liquid effluent monitoring device should be sufficiently sensitive to beta radiation for measuring concentrations of 1×10^{-7} $\mu\text{Ci}/\text{ml}$ with reasonable speed and accuracy. Monitoring instruments employing immersible beta sensitive

FIGURE 8. A Detector Probe for Gamma Monitor

FIGURE 9. A Readout Meter for Gamma Monitor

detectors are commercially available and in use, but none are sufficiently sensitive to measure $1 \times 10^{-7} \mu\text{Ci/ml}$ of $\text{Sr}^{90}\text{Y}^{90}$. Seuba and Veburg (1959) described an immersion type GM tube having thin walls which in 2 in. of lead shielding requires a concentration of $2 \times 10^{-6} \mu\text{Ci/ml}$ to produce a net count equal to background.

Several problems are inherent in the use of immersed detectors. One vexsome problem is that the fluid

being monitored forms part of the background shielding, and accurate background determination is not possible when the plant is operating. When detectors are located in low background areas, variable radiation levels from adjacent equipment and piping can be a problem. Also activity tends to buildup on the surface of the detector due to adsorption and, in some cases, algae growth and the reliability of background counting is

adversely affected. Therefore, it is necessary to design detectors that can be easily removed from the line for cleaning and decontamination.

Continuous Sampling and Counting

By removing a volume from the effluent stream for counting, it is possible to optimize the geometry of the detector, the shielding, and to provide for anticoincidence counting. An example of the degree of possible improvement can be seen by comparing the obtainable natural background rates with a GM tube expressed as residual percentages of the initial unshielded background

5 cm lead	33%
20 cm lead	25%
20 cm lead anticoincidence	1 to 2%.

The effect of shielding is even greater where there are sources of radiation near a monitor.

Smith reports a device using a sample holding vessel over which a detector is fitted (Figures 10 and 11). The contents of the sample holding vessel can be changed on demand or continuously, and are automatically drained and flushed periodically. The background is checked during this period.

An apparatus for the continuous monitoring of beta-gamma activity level in a process water stream at Savannah River is described by Wingfield and Liller. A hollow cylinder of water is established which surrounds, but does not touch, a plastic scintillator. The wall of water is maintained by the momentum of the influent, which is tangential to the inner cell wall. There is no buildup of activity on the detector

because it is not touched by the solution being counted. The device can reliably measure concentrations of 10^{-5} $\mu\text{Ci/ml}$.

Concentration of the Radionuclides in a Sample

If the radionuclides in a sample can be concentrated, the problem involved in reliably counting their radioactivity are greatly diminished. Three methods of concentration are normally used in radiochemistry, i.e., evaporation, chemical precipitation/flocculation, and ion exchange and solvent extraction. Chemical precipitation and flocculation would seem to have little application to the monitoring of the very pure water constituting power reactor wastes. Further, if beta counting is required, the field is narrowed to evaporation.

Several very interesting automatic monitoring devices, which concentrate samples of water for counting, have been developed and provide interest-

FIGURE 10. Liquid Sampling Cell

FIGURE 11. Schematic of Continuous Effluent Monitor

ing background information even though they are not strictly applicable to the conditions at a power reactor plant.

Rieck described a device for monitoring the concentration of As^{76} in Hanford reactor cooling water. Gamma energy measurements are made with a sodium iodide crystal on samples automatically pretreated to remove interfering nuclides by cation exchange resins and a 3 hr decay. The counting arrangement is shown in Figure 12. The minimum detection of this device is 2×10^{-5} μCi of As^{76} ml of reactor cooling water.

The As^{76} monitor was preceded at Hanford by the development of a device

FIGURE 12. Monitoring Device, Gamma Energy Measurements Made with a NaI Crystal

that continuously analyzed the total activity of a sample of reactor effluent water and also the concentration of the seven prominent radio-nuclides. Rieck, Ratcliffe, and Schwendiman describe the instrument (Figure 13) that meters sample volumes into a cup, moves some of the cups through a drying operation to a counting position, removes interfering isotopes from some of the sample

by chemical methods, counts for selected nuclides, and records the results. The particular nuclides selected were Na^{24} , As^{76} , Np^{239} , Mn^{56} , Cu^{64} , Si^{31} , and P^{32} .

Gaseous Wastes

All potentially contaminated gaseous wastes are released via stacks designed to reduce airborne concentrations at ground level to an acceptable level during some maximum emergency condition.

If the concentrations of radionuclides in the stack gases routinely released are expected to be about 1/10 or more of the safety limits, there is an advantage to monitoring the gases in the stack after dilution with uncontaminated air streams which also discharge through the stack. Monitoring the gases in the stack will undoubtedly be a condition of AEC operating approval in such plants. If the concentrations in a stack are expected to be less than about 1/10 of the safety limits, monitoring the radioactive gases upstream of the stack, when they are as concentrated as possible, gives the advantage of increased convenience, reliability, and sampling accuracy.

An accurate measurement of the radioactive content of gaseous wastes requires that the radionuclides in both particulate and gaseous form be included in the measurement. If a sample of the waste must be collected for analysis, as is generally the case, different techniques must be used to collect the solids and the gases. A fact probably not important in present reactor practice but which should be noted is that some fission products do change physical states while in transport and during collection. The halides, notably iodine, can exist as a chemically pure condensable vapor or as a contaminant associated either chemically or physically with other solids, liquids or gases.

Chemically inert gases such as argon, krypton, and xenon are easily sampled and transported via pipes, but are difficult to accumulate and concentrate for analysis. Particulates

do not transport as easily, and truly representative samples require special equipment.

The lower limit of detection of a gas monitoring system is limited by the "background" radiation. It can be extended by:

- Using shielding to decrease the original background radiation level
- Relocating of the detector to a place with a lower level of background radiation
- Increasing the volume of gas counted
- Increasing the counting period.

Inert Gas Monitoring

The noble gases, e.g., Ar⁴¹, Kr⁸⁵, and Xe¹³³, are difficult to measure at low concentrations. They are chemically inert, and can only be concentrated by means of a freeze trap or a charcoal sampler maintained at a very low temperature. However, these techniques are not suitable for routine operation, and direct measurement of the gas is the only suitable technique.

GM tubes, scintillation crystals, and ionization chambers can be used to measure the activity of the noble gases contained in chambers of known volumes. The gas to be placed in the chamber must first be cleansed of particulates and reactive gases so that the contents of the chamber can be assumed to be only noble gases and free of contamination.

An ionization chamber for the direct monitoring of the effluent coolant from gas cooled reactors was developed by Kanne. It is a large volume folded flow chamber, adapted to batch or continuous analysis and primarily sensitive to beta emissions. With a sensitivity on the order of 5×10^{-13} A,

this chamber will detect noble gas concentrations on the order of $10^{-7} \mu\text{Ci}/\text{cm}^3$. The main disadvantages of the Kanne chamber are those associated with the qualitative aspects of the measurement and the need to periodically purge the chamber to decontaminate it. In a free-flow system, all contaminants are cumulatively measured with no means for differentiating among them. Soldat (1962) reported that a tritium sampler (Kanne chamber) with a filter and charcoal trap ahead of it was successfully used to measure K^{85} concentrations down to approximately $2 \times 10^{-7} \mu\text{Ci}/\text{cm}^3$. GM tubes and scintillation crystals can be used to measure radioactive gases by mounting a detector in a shielded chamber and filling the chamber with the gas either batchwise or continuously. Typically, a concentration of about $10^{-6} \mu\text{Ci}/\text{cm}^3$ of Ar^{41} will produce a count rate equal to background in such a chamber.

Examples of Gas Monitors

Dresden Nuclear Power Station

The original off-gas monitor was a gas sampling and measuring device which used a $\text{NaI}(\text{Tl})$ crystal to count a 0.68 ft^3 volume of gas. This monitor was replaced by an ion chamber device early in 1965 after the ion chamber response had shown superior correlation with known core conditions. A block diagram of the ion chamber device is shown (Figure 14).

Yankee-Rowe

The air ejector monitor at the Yankee plant is a cylindrical anthracene crystal (1 1/2 in. in diam and 1/8 in. thick) mounted in a chamber shielded by 2 in. of lead (Figures 15-16). The crystal is sealed by an

FIGURE 14. Block Diagram of Dresden Off-Gas Monitor

aluminized mylar foil having a thickness of less than $2 \text{ mg}/\text{cm}^2$. The chamber is located 15 ft downstream of the air ejectors. Strictly speaking, the air ejector monitor is a process monitor rather than an effluent monitor, because the air ejector exhaust gases are temporarily retained in tanks before being released.

FIGURE 15. Yankee-Rowe Air Ejector Monitor Detector

FIGURE 16. Yankee-Rowe Air Ejector Monitor Instrument Panel

The detector output is indicated in counts per minute on a log ratemeter in the control room. The ratemeter time constants vary from 72 sec at 10 counts/min to 0.012 sec at 10^6 counts/min. The monitor was originally calibrated against a known concentration of Kr^{85} in nitrogen gas. The calibration is checked monthly by compar-

ing the response to a Co^{60} source with previous response. Accuracy is reported as $\pm 15\%$ of actual count rate on a three decade scale and $\pm 25\%$ on a five decade scale. Maximum sensitivity is reported to be $2 \times 10^{-7} \mu\text{Ci}/\text{cm}^3$; but tests have shown a sensitivity of $\sim 10^{-6} \mu\text{Ci}/\text{cm}^3$.

APPENDIX A

COMPLIANCE WITH REGULATORY REQUIREMENTSRegulatory Authorities

The Atomic Energy Act of 1954 authorized the U.S. Atomic Energy Commission to license the use of nuclear and radioactive by-products materials by qualified applicants pursuant to such safety standards as established by rule of the Commission to protect health and to minimize danger to life and property. Prior to that time, the users of such materials had worked under a contract with the USAEC and were required to comply with the radiation protection requirements of the AEC Manual. With licensing authority came the need for a modified presentation of the radiation protection requirements.

Accordingly, licensing standards were promulgated and published in the Code of Federal Regulations under Title 10 - Atomic Energy. The standards of principal radiation protection interest are Part 20 - STANDARDS FOR PROTECTION AGAINST RADIATION and Part 100 - REACTOR SITE CRITERIA. These published standards serve as the minimum requirements with which licensee's must comply, and they are often augmented by further requirements imposed as a condition of license approval.

Until 1959, the federal government was the sole control authority for the use of radioactive materials within the United States. (Certain naturally occurring radioactive materials, such

as radium; and radiation producing machines, such as X-ray devices, were excluded from federal control.) In 1959, an amendment to the Atomic Energy Act of 1954 made possible the transfer of control authority from the federal government to qualified state governments. At the time of this writing, such control authority had been transferred to nine states, viz., Kentucky, Mississippi, California, Kansas, New York, Arkansas, Texas, North Carolina, and Florida. The procedures leading to transfer of authority were in various stages of completion with several other states. It should be noted that the 1959 amendment to the Atomic Energy Act authorized transfer of control authority for

- by-product materials
- source materials
- special nuclear materials in quantities not sufficient to form a critical mass.

Thus, the authority to control reactors, which require a critical mass of special nuclear material, is not contained in the amendment and is, therefore, retained in the federal government.

Also in 1959, the formation of a Federal Radiation Council was authorized by Congress to " . . . advise the President . . . with respect to radiation matters directly or indirectly related to health." The

Council was instructed to establish a national policy related to radiation exposure. Since its formation the FRC has issued eight reports for the purpose of providing radiation protection guidance" . . . for all Federal agencies on the formulation of radiation standards and in the establishment and execution of programs of cooperation with States." Thus, it is not entirely accurate to categorize the FRC as a regulatory authority, but the error in doing so in this discussion is not serious, and the FRC does exert an influence of the standards developed by the truly regulatory authorities.

The role of public health agencies in radiation protection has been the subject of considerable discussion. It is probably sufficient for the purposes of this document to acknowledge that public health agencies from local to federal level are charged with the responsibility of protecting the health and well-being of the citizenry. Several sources of radiation exposure, especially fallout from atomic weapons testing, have been extensively measured by these agencies.

Regulations

The recommendations of the International Commission on Radiological Protection, the National Commission on Radiological Protection, and the Federal Radiation Protection Council form the basis of nearly all current radiation protection regulations. A discussion of these regulations is essential to a comprehensive review of regulations. A word of caution concerning "summaries" and "digests" of the findings and recommendations of these

groups is appropriate at this point. The three organizations are careful to explain at length the philosophy and intent underlying their work and to describe the manner in which their findings should be applied. Unless a person is familiar with these explanations, the recommendations when reported out of context can easily be misconstrued. Thus, it behooves a person with radiation protection responsibility to become acquainted with the reports of the ICRP, NCRP and FRC.

The recommendations of the ICRP and NCRP are primarily directed toward the protection of occupationally exposed persons. However, recent recommendations do include statements applicable to persons outside of controlled areas. In general, the recommendations pertaining to persons outside of controlled areas specify a maximum radiation dose or rate of intake equivalent to 1/10 of that dose or rate of intake permitted for occupational exposure. These recommendations include maximum permissible concentrations (MPC's) of radioactive contamination in air; in effect, secondary standards - the primary standards being the maximum permissible dose. The recommendations are not reproduced in part in this document because the recommendations should be consulted in toto to be meaningful, and their entire reproduction would be too ambitious for this report. However, a discussion of the pertinence of the recommendations to effluent monitoring and environmental should serve some useful purpose.

Neither ICRP nor NCRP has given specific guidance regarding the contamination of food or environmental materials other than air and water. However, several general comments relating to environmental and food contamination do appear in the main ICRP recommendations and in the 1959 Report of Committee II on Permissible Dose for Internal Radiation.

"Members of the population at large may be exposed to radiation that cannot be related to any specific controlled area . . . As such exposure is not easily controlled, it will be impossible to ensure that a recommended maximum permissible individual dose is not exceeded in any single case. Where large numbers are involved, it will not be possible to examine the habits of every individual. A reasonable procedure would be to study a sample of the group involved and to set the environmental level so that no individual in the sample receives any excessive exposure. There will always remain the possibility that someone of grossly different habits from those in the observed sample may receive a higher dose than the maximum in the sample."

While the above quotation is more applicable to the establishment of maximum permissible levels than to a monitoring program, it does indicate that the habits and customs of people do affect the degree of exposure to radioactive materials in the environment. It also theorizes that the radiation exposure of a population can be determined by measuring the exposure of a representative sample of that population.

AEC Licensing Regulations

Three sections of 10 CFR Part 20, Standards for Protection Against Radiation, are concerned with the release of radioactive materials:

Concentrations in Effluents to Unrestricted Areas

- 1) There may be included in any application for a license, or for amendment of a license, proposed limits upon concentrations of licensed and other radioactive material released into air or water in unrestricted areas as a result of the applicant's proposed activities. Such applications should include information as to anticipated occupancy times for each unrestricted area involved. The Commission will approve the proposed limits if the applicant demonstrates that it is not likely that any individual will be exposed to concentrations in excess of the limits specified in Appendix B, Table II, of this part.* For purposes of this paragraph, concentrations may be averaged over periods not greater than one year.
- 2) Except as authorized by the Commission pursuant to 20.302 or paragraph (2) of this section, no licensee shall possess, use or transfer licensed material in such a manner as to release into air or water in any unrestricted area any concentration of radioactive material in excess of the limits specified in Appendix B, Table II, of this part. For purposes of this paragraph, concentrations may be averaged over periods not greater than one year.
- 3) For purposes of this section, determinations as to the concentration of radioactive material shall be made with respect to the point where such material leaves the restricted area. Where the radioactive material is discharged through a stack, tube, pipe, or similar conduit, the determination may be made with respect to the point where the material leaves such conduit.
- 4) The provisions of this section do not apply to disposal of radioactive material into sanitary sewerage systems (see 20.303).

* Note: These concentrations are 1/10 of the 168 hour occupational MPC determined by the NCRP.

Methods for Obtaining Approval of Proposed Disposal Procedures

Any licensee or applicant for license may apply to the Commission for approval of proposed procedures to dispose of licensed material in a manner not otherwise authorized in the regulations in this chapter. Each application should include a description of the licensed material and any other radioactive material involved, including the quantities and kinds of such material and the levels of radioactivity involved, and the proposed manner and conditions of disposal. The application should also include an analysis and evaluation of pertinent information as to the nature or the environment, including topographical, geological, meteorological, and hydrological characteristics; usage of ground and surface waters in the general area; the nature and location of other potentially affected facilities; and procedures to be observed to minimize the risk of unexpected or hazardous exposures. The Commission will not approve any application for a license to receive licensed material from other persons for disposal on land not owned by the Federal government or by a state government.

Disposal by Release into Sanitary Sewerage Systems

No licensee shall discharge licensed material into a sanitary sewerage system unless:

- 1) It is readily soluble or dispersible in water; and
- 2) The quantity of any licensed or other radioactive material released into the system by the licensee in any one day does not exceed the larger of subparagraphs (a) or (b) of this paragraph:
 - a) The quantity which, if diluted by the average daily quantity of sewage released into the sewer by the licensee, will result in an average concentration equal to the limits specified in Appendix B, Table I, Column 2 of this part;* or
 - b) Ten times the quantity of such material specified in Appendix C of this part; and
- 3) The quantity of any licensed or other radioactive material released in any one month, if diluted by the average monthly quantity of water released by the licensee, will not result in an average concentration exceeding the limits specified in Appendix B, Table M, Column 2 of this part; and
- 4) The gross quantity of licensed and other radioactive material released into the sewerage system by the licensee does not exceed one curie/yr.

Excreta from individual undergoing medical diagnosis or therapy with radioactive material shall be exempt from any limitations contained in this section.

AEC MANUAL

The radiation protection requirements for direct contract work with the AEC, as specified in the AEC Manual, and the licensee requirements (10 CFR Part 20) are technically identical. Some differences do occur in the interpretation and application of the requirements, however. In general, the maximum permissible concentrations are applied at the "point of discharge" in the case of the licensee and at the "point of use" exposure evaluations not required of a licensee. Further, the exposures in "uncontrolled areas" must not exceed certain standards suggested by the Federal Radiation Council, viz,

* *Note: These concentrations are equal to the 40 hour occupational MPC determined by the NCRP*

Radiation Protection Standards for Individuals and Population Groups
in Uncontrolled Areas

Radiation Dose Standards for External and Internal Exposure

<u>Type of Exposure</u>	<u>Dose (rem/yr)</u>	
	<u>Based on Exposure to Individuals</u>	<u>Based on an Average Exposure to a Suitable Population Sample</u>
Whole Body, Gonads, or Bone Marrow	0.5	0.17
Thyroid or Bone	1.5	0.5
Bone (alternate standard)	Body burden of 0.003 μg of Ra^{226} or its biological equivalent	Body burden of 0.001 μg of Ra^{226} or its biological equivalent

Radioactivity in Effluents Released
To Uncontrolled Areas

- 1) Except as provided in paragraph 2 below of this section, radioactivity in effluents released to uncontrolled areas shall not exceed the radiation protection standards specified in Table II of Annex 1 of this appendix. The point of release of such effluents shall be considered to be the point at which the effluents pass beyond the site boundary. Where such effluents are discharged through a conduit, such as a stack or pipe, the point of release may be considered to be the conduit discharge. For the purpose of applying these standards, radioactivity concentrations in effluents may be averaged over periods up to one yr.
- 2) Radioactivity in effluents may be released to uncontrolled areas in excess of the radiation protection standards specified in Table II of Annex 1 of this appendix provide-

ed it is reasonably demonstrated that in uncontrolled areas:

- a) individuals are not exposed in excess of the standards specified in paragraph A, above.
- b) individuals are not exposed in excess of Table II of Annex 1 standards, or
- c) the average exposure of a suitable sample of an exposed population group is not in excess of 1/3 of Table II of Annex 1 standards. Radioactivity concentrations in the environment may be averaged over periods up to one year.

Federal Radiation Council (FRC)
Recommendations

The Federal Radiation Council (FRC), a regulatory organization formed in 1959, has given more attention to the need for environmental surveillance than have either the ICRP or the NCRP. The first staff report of the FRC introduced the term "Radiation Protection Guide" (RPG) viz.

"Radiation Protection Guide (RPG) is the radiation dose which should not be exceeded without careful consideration of the reasons for doing so; every effort should be made to encourage the maintenance

of radiation doses as far below this guide as practicable."

The first staff report also suggests and operational technique for assuring that the RPG for the annual whole body dose to the individual in the population is not exceeded when the actual dose received by each individual is not measured. The technique is to measure the dose received by a suitable sample of the population and to impose an RPG on the average of the sample, which is 1/3 of the RPG imposed on individuals in the general population.

Further guidance on the control of environmental radioactivity is given by the second report of the Federal Radiation Council which discusses that subject in more detail than have any previous advisory or regulatory reports, viz.,

"The objective of the control of population exposure from radio-nuclides occurring in the environment is to assure that appropriate RPG's are not exceeded. This con-

trol is accomplished in general either by restriction on the entry of radioactive materials into the environment or through measures designed to limit the intake of such materials by members of the population. The most direct means of evaluating the effectiveness of control measures is the determination of the amount of radioactive material in the bodies of the members of exposed population groups. Although the determination of such body burdens may at times be indicated, in routine practice potential exposures will generally be assessed on the basis of either one or a combination of two general approaches: (1) calculations based upon known amounts of radioactive material released to the environment, and assumptions as to the fraction of this material reaching exposed population groups, or (2) environmental media."

The second report also recommended a graded series of radiation protection actions which are related to ranges of intake of radioactive materials by exposed population groups, viz.,

<u>Ranges of Transient Rates of Daily Intake</u>	<u>Graded Scale of Action</u>
Range I	Periodic confirmatory surveillance as necessary
Range II	Quantitative surveillance and routine control
Range III	Evaluation and application of additional control measures as necessary

State Regulations

Since the authority for the regulation of the use of special nuclear materials was not altered by the 1959 amendment to the Atomic Energy Act of 1954, there would seem to be no reason to discuss state regulations in this document. The actual authority of a state to regulate the actions of a power

reactor are still somewhat in doubt, however. In the absence of clearly defined authority, agreements have been worked out between several power reactor plant operators and state agencies, especially the departments of public health and water pollution control authorities.

APPENDIX B

IDENTIFICATION OF RADIONUCLIDE
COMPOSITION OF POWER REACTOR WASTESIntroduction

A reactor produces radionuclides through fission of the fuel, activation of material exposed to the neutron field, and transmutation of fertile and fissionable materials. The bulk of the radionuclides are the fission products that remain intimately associated with the fuel material. They remain with the fuel until removed by processing at a fuel processing plant. Since this guide is principally concerned with the power reactors, the bulk of the fission products intended for shipment to processing plants can be ignored. The important radionuclides in this discussion are (1) the minute fractions of fission products that do not remain associated with the fuel and (2) the activation products occurring in various reactor systems. Practically all of these materials will appear ultimately as radioactive waste which must either be contained or dispersed.

The relative yields of the individual fission products have been well described in the literature. Culler (1959) listed the most significant, classified by half-lives and yield as shown in Table B-I.

The composition of radionuclides formed by neutron activation differs according to the structural and coolant materials used in the various types of reactors. Table B-II summarizes the

typical activation products in power reactor systems.

Water is the most used cooling medium in U.S. power reactor design. During the operation of water-cooled reactors, radionuclides are produced by activation of the water by neutrons and protons (Table B-III) and by activation of impurities in the water. The principal radionuclides of interest are shown in Table B-IV.

The composition of the radionuclides in power reactor wastes is affected not only by the radionuclides generated but also by a variety of plant characteristics. One example is that the closed primary loop of a pressurized water reactor allows short lived radionuclides to decay before they appear as a waste. The more important characteristics affecting the nature and amount of radioactive wastes at 11 power reactor stations, are listed in Table B-V and described below.

PRESSURIZED WATER REACTORS

The Shippingport, Yankee, and Indian Point reactors use a pressurized water cycle as illustrated in Figure B-1 (Sketch A). In this cycle the primary cooling water circulates under high pressure (1800 to 2500 psia) and as in any circulating water system, some corrosion of the surfaces in contact with the coolant occurs. Most of the

surface exposed to the primary coolant in these reactors is stainless steel. Therefore, the corrosion products are constituents of stainless steel materials. Two sources of activated corrosion products exist in the primary system. The more important is the surface area located in regions of high neutron flux, such as the fuel cladding. In addition, the surfaces located outside the high flux region,

such as the primary coolant piping, steam generators and pumps contribute corrosion products which become radioactive as they pass through the high flux region of the core. A corrosion rate estimate of 10 mg/meter²/mo is typical for a pressurized water reactor.

Defects in the fuel cladding allow fission products to escape to the primary coolant where they accumulate. The design of auxiliary systems is

TABLE B-1. Principal Fission Products of Interest in Waste Disposal Operations^(a)

Radionuclides	Half-Life (b)		Approximate Percent of Total Activity Due to Specific Isotopes After the Following Decay Times (c)		
	Days	Years	100 Days	3 Years	30 Years
Sr-Y ⁹⁰		28.0	<2%	15%	~49%
Cs-Ba ¹³⁷		26.6	<2	15	~49
Pm ¹⁴⁷		2.6	3	15	<1
Cr-Pr ¹⁴⁴	290.0		45	50	-
Kr ⁸⁵		10.3	<1	1	<1
I ¹³¹	8.1		<1	-	-
Zr-Nb ⁹⁵	63.0		33	-	-
Ba-La ¹⁴⁰	12.8		<1	-	-
Ru-Rh ¹⁰³	41.0		5	-	-
Ru-Rh ¹⁰⁶		1.0	2	3	-
Sr ⁸⁹	54.0		7	-	-
Xe ¹³³ (gas)	5.3		<1	-	-
			100%	100%	100%

(a) Adapted from "Nature of Radioactive Wastes," testimony presented by F. L. Culler, Jr., before the Joint Committee on Atomic Energy Hearings on "Industrial Radioactive Waste Disposal," 1959, vol. 1, p. 22.

(b) The time during which half of a given amount of material decays:
d = days; y = years

(c) The sign "<" means "less than"; the sign "~" means "approximately."

TABLE B-II. Typical Activation Products Which Appear in Wastes from Power Reactors^(a)

Radionuclide ^b	Half-Life ^c	Mechanisms of Formation ^d	Activated Source
Light-Water-Cooled Reactors:			
Nitrogen-16	7 s	O-16 (n, p) N-16	water
Nitrogen-17	4 s	O-17 (n, p) N-17	water
Oxygen-19	30 s	O-18 (n, p) O-19	water
Hydrogen-3 (Tritium) ^(e)	12.0 y	Li-6 (n, α) H-3	lithium hydroxide
Fluorine-18	1.8 h	O-18 (p, n) F-18	water
Sodium-24	15.0 h	Na-23 (n, γ) Na-24	sodium in water
		Al-27 (n, α) Na-24	aluminum
Aluminum-28	2.0 m	Al-27 (n, γ) Al-28	aluminum
Silicon-31	2.8 h	P-31 (n, p) Si-31	water treatment with phosphates
Argon-41	1.8 h	A-40 (n, γ) A-41	air
Manganese-54	300 d	Fe-54 (n, p) Mn-54	steel
Manganese-56	2.6 h	Mn-55 (n, γ) Mn-56	steel
		Fe-56 (n, p) Mn-56	steel
Iron-55*	2.9 y	Fe-54 (n, γ) Fe-55	steel
Iron-59*	45.0 d	Fe-58 (n, γ) Fe-59	steel
		Co-59 (n, p) Fe-59	steel
Cobalt-58*	71.0 d	Co-57 (n, γ) Co-58	steel and Stellite
		Ni-58 (n, p) Co-58	steel
Cobalt-60*	5.2 y	Co-59 (n, γ) Co-60	steel
Copper-64	12.8 h	Cu-63 (n, γ) Cu-64	{ 17-4 ph steel and Cu-Ni alloy
Copper-66	4.6 m	Cu-65 (n, γ) Cu-66	
Nickel-65	2.6 h	Ni-64 (n, γ) Ni-65	steel
Chromium-51*	27.0 d	Cr-50 (n, γ) Cr-51	steel
Zirconium-95	65 d	Zr-94 (n, γ) Zr-95	zirconium fuel structure
Hafnium-181	40 d	Hf-180 (n, γ) Hf-181	hafnium control rods
Tantalum-182*	111.0 d	Ta-181 (n, γ) Ta-182	steel
Tungsten-187	24.0 h	W-186 (n, γ) W-187	Stellite
Sodium-Cooled Reactors:			
Argon-41	1.8 h	A-40 (n, γ) A-41	impurity in nitrogen
Sodium-24*	15.0 h	Na-23 (n, γ) Na-24	sodium
Sodium-22*	2.6 y	Na-23 (n, 2n) Na-22	sodium
Rubidium-86*	19.5 d	Rb-85 (n, γ) Rb-86	coolant impurities
Antimony-124*	60.0 d	I-127 (n, α) Sb-124	coolant impurities

^a Derived from testimony presented at the 1959 Congressional hearings on "Industrial Radioactive Waste Disposal," by F. L. Culler and F. R. Bruce, as well as operating experience on various reactors.

^b The numbers following element names are isotopic mass numbers.

^c The time during which half of a given amount of material decays: s = seconds; m = minutes; h = hours; d = days; y = years.

^d For example, O-16 (n, p) N-16 means that nitrogen-16 is formed from the naturally occurring isotope oxygen-16 by the absorption of a neutron (n) and the ejection of a proton (p). α and γ refer to alpha particles and gamma rays, respectively. (See "Types of Radiation" in Section II of this report.)

^e Asterisks indicate the isotopes likely to be most prevalent and which can be important in waste handling operations.

TABLE B-III. Water Activation Products

Isotope	Nuclear Reaction	Half-Life	Beta		Position		Gamma		Total Decay Energy, MeV
			Energy, MeV	Quantity, %	Energy, MeV	Quantity, %	Energy, MeV	Quantity, %	
H ³ (a)	H ² (n,γ), Li ⁶ (n,α) (b)	12.26 yr	0.0186	100					0.0186
N ¹³	O ¹⁶ (p,α)	10.0 min			1.19	100			2.21
N ¹⁶	O ¹⁶ (n,p) N ¹⁵ (n,γ)	7.4 sec	10.4 4.3	26 68			6.13 7.12	73 5	10.4
N ¹⁷	O ¹⁷ (n,p)	4.14 sec	3.7						8.8
O ¹⁹	O ¹⁸ (n,γ)	29 sec	3.25 4.60	58 42			0.200 1.36	96 54	4.80
F ¹⁸	O ¹⁸ (p,n)	1.87 hr	(c)	3	0.65	97			1.67
Ar ³⁷ (d)	Ar ³⁶ (n,γ)	35 days	(c)	100					0.82
Ar ⁴¹ (d)	Ar ⁴⁰ (n,γ)	1.83 hr	1.20 2.49	99 1			1.29	99	2.49

(a) Tritium; although tritium is a gas, it is usually present in more than trace quantities only in liquid wastes discharged from heavy-water-cooled reactors. However, measurable concentrations have been observed in the coolant of several power reactors, e.g., Yankee-Rowe, where it is carried over to the distillate during waste treatment by evaporation thus appearing in the liquid waste discharged from the plant.⁷

(b) Lithium is sometimes added to coolants for pl¹ control; LiH⁷ is used at Shippingport.

(c) Electron capture; gamma rays characteristic of the daughter are emitted

(d) Argon -37 and -41 may build up in small amounts if the water is not deaerated.

TABLE B-IV. Impurity Activities in Water ^(a)TABLE B-V. Plant Characteristics Which Affect Radioactive Waste Generation ^(a)

Isotope	Nuclear Reaction	Half-Life	Energy and Quantity of Most Significant Decay Particles						Total Decay Energy, MeV
			Beta		Position		Gamma		
			Energy, MeV	Quantity, %	Energy, MeV	Quantity, %	Energy, MeV	Quantity, %	
Na ²⁴	Al ²⁷ (n,α), Na ²³ (n,γ)	15 hr	1.39	100			2.75 1.37	100 100	5.51
Al ²⁸	Al ²⁷ (n,γ)	2.30 min	2.87	100			1.78	100	4.65
Si ³¹	P ³¹ (n,p)	2.62 hr	1.48						1.48
Cl ³⁸	Cl ³⁷ (n,γ)	37.3 min	4.8 1.1	53 31			2.1 1.6	47 31	4.8
Ar ³⁷	Ar ³⁶ (n,γ)	35 days	(a)	100					0.82
Ar ⁴¹	Ar ⁴⁰ (n,γ)	1.83 hr	1.20 2.49	99 1			1.29	99	2.49
Ca ⁴⁵	Ca ⁴⁴ (n,γ)	160 days	0.25	100					0.25
Cr ⁵¹	Cr ⁵⁰ (n,γ)	27.8 days	(a)				0.32	10	0.75
Mn ⁵⁴	Fe ⁵⁴ (n,p)	280 days	(a)	100			0.84	100	1.38
Fe ⁵⁵	Fe ⁵⁴ (n,γ)	2.6 yr	(a)	100					0.22
Mn ⁵⁶	Fe ⁵⁶ (n,p), Mn ⁵⁵ (n,γ)	2.58 hr	2.86 1.05	60 24			0.84 1.81 2.11	99 23 14	3.70 2.31 1.56
Co ⁵⁸	Ni ⁵⁸ (n,p)	71 days	(a)	85	0.48	15	0.81 1.62	100 0.5	2.31
Fe ⁵⁹	Co ⁵⁹ (n,p)	45 days	0.46 0.27	53 46			1.10 1.29	56 44	1.56
Co ⁶⁰	Co ⁵⁹ (n,γ)	5.27 yr	0.31	100			1.33 1.17	100 100	2.81
Cu ⁶⁴	Cu ⁶³ (n,γ)	12.9 hr	0.57 (a)	38 43	0.66	19	1.34	1	(b)
Ni ⁶⁵	Ni ⁶⁴ (n,γ)	2.56 hr	2.10 0.6 1.01	69 23 8			1.49 1.11	20 10	2.10
Zn ⁶⁵	Zn ⁶⁴ (n,γ)	245 days	(a)	98	0.33	2	1.11	50	1.35
Cu ⁶⁶	Cu ⁶⁵ (n,γ)	5.1 min	2.63 1.59	91 9			1.04	9	2.63
Sr ⁸⁹	Sr ⁸⁸ (n,γ)	51 days	1.46	100					1.46
Zr ⁹⁵	Zr ⁹⁴ (n,γ)	65 days	0.40 0.36	50 43			0.72 0.76	55 43	1.12
Hf ¹⁸¹	Hf ¹⁸⁰ (n,γ)	45 days	(c) 0.41				0.137		1.02
Ta ¹⁸²	Ta ¹⁸¹ (n,γ)	115 days	0.51	8			0.10 1.12 1.22 1.19	56 33 28 15	1.73
Ta ¹⁸³	Ta ¹⁸² (n,γ)	5.0 days	0.61	91			0.108 0.246	44 35	1.07
W ¹⁸⁷	W ¹⁸⁶ (n,γ)	24.0 hr	0.63	70			0.69 0.48 0.134		1.31

(a) W. A. Rodger. "Safety Problems Associated with the Disposal of Radioactive Waste," *Nuclear Safety*, vol. 5, number 4, p. 293. Summer, 1964.

(b) Electron capture

(c) Electron capture and positron, 1.68; beta, 0.57.

(d) Conversion electron

TABLE B-V. Plant Characteristics Which Affect Radioactive Waste Generation

	SHIPPING- PORT	YANKEE	INDIAN POINT	DRESDEN	ENRICO FERMI	HALLAM
Coolant	Light water	Light water	Light water	Light water	Sodium	Sodium
Coolant Cycle	Pressurized water	Pressurized water	Pressurized water	Direct dual-cycle boiling water	Sodium-to-water	Sodium-to-water
Type of Fuel	Seed: 7% U-93% Zr alloy plates clad in Zr-2. Blanket: UO ₂ ceramic pellets in Zr-2 tubes	UO ₂ ceramic pellets in stainless steel tubes	UO ₂ -ThO ₂ ceramic pellets in Zr-2 tubes	UO ₂ ceramic pellets in Zr-2 tubes	Seed: 90% U-10% Mo metal rods clad in Zr. Blanket: U metal rods clad in stainless steel	90% U-10% Mo metal clad in stainless steel. (Second core will be clad uranium carbide)
Maximum Fuel Failures ^b	1% ^c	1% ^c	0.5% ^c	1.4% ^c	0.1% ^d	1% ^c
Control Rod Materials	Hafnium	Silver-indium-cadmium alloy clad with nickel	Hafnium	2% boron-stainless steel	B ₄ C in stainless steel tubes	Clad rare earth oxides
Other Material Exposed to Primary Coolant:						
Austenitic stainless steel (primary equipment)	Yes	Yes	Yes	Yes	Yes	Yes
Zircaloy-2 (fuel structure)	Yes	No	Yes	Yes	Zirconium	Yes
Stellite (valve seats)	Yes	Yes	Yes	Yes	Yes	Yes
Other	No	No	No	Yes ^e	Yes	Yes ^f
Coolant Purification	Full-pressure by-pass filtration and ion exchange	Low-pressure by-pass ion exchange	Low-pressure by-pass filtration, ion exchange, degasification	By-pass demineralization (primary feed gets full-flow demineralization)	By-pass sodium cold traps	By-pass sodium cold traps and carbon hot traps
Additives to Coolant:						
During operation	H ₂ and LiOH	H ₂	H ₂	Morphaline, NaOH and sodium phosphate added to secondary circuit feed water only	No	No
During cold shutdown	No	Soluble boron	Soluble boron	No	No	No
Maximum Number of Cold Shutdowns per Year ^b	2	2	3	2	1 ^g	1 ^g

^a Stainless steel cladding is being considered.

^b Maximum design estimate basis.

^c Percent of fuel envelopes with pinhole-type clad failures.

^d Percent of active fuel surface exposed to coolant.

^e Primary steam pipe is 2¼% Cr-1% Mo steel; 20,000 sq ft of feedwater heater surface is Monel and 20,000 sq ft is 70% Cu-30% Ni; main condenser tubes are Admiralty metal; there are also special turbine materials.

^f Some 405 stainless is used.

^g Shut down to 500 F or lower.

(a) Reproduced with permission from "Radioactive Waste Handling In The Nuclear Power Industry," A Report of the Technical Appraisal Task Force on Nuclear Power to the Board of Directors of the Edison Electric Institute, March, 1960.

PRESSURIZED WATER CYCLE

Sketch A

DIRECT DUAL BOILING WATER CYCLE

Sketch B

SODIUM CYCLE

Sketch C

FIGURE B-1. Sketch of Power Reactor Coolant Loops^(a)

(a) Reproduced with permission from "Radioactive Waste Handling in the Nuclear Power Industry," Edison Electric Institute, March, 1960.

usually based on the supposition that 1% of the fuel rods will develop minor defects such as minute cracks and pinholes.

Table B-VI lists the radioactive isotopes in the primary coolant of the Yankee reactor that have an equilibrium level greater than 0.01 $\mu\text{Ci/ml}$. These equilibrium activities assume a constant 10 gpm flow from the primary system to the purification system. The volume of

gas occurring in the primary coolant is relatively low. Hydrogen is the principal gas waste although fission gases, especially Xe^{133} and Kr^{85} , can be present due to fuel element failures. These gases come out of solution when the primary cooling water is depressurized. They appear in the secondary system only if there are leaks in the main steam generator. In this case, radioactive gases would appear in the

TABLE B-VI. Design Estimates of Radionuclides in Yankee Primary Coolant

Nonvolatile Fission Products		Volatile Fission Products	
Isotopes	Activity, $\mu\text{Ci/ml}$	Isotopes	Activity, $\mu\text{Ci/ml}$ (liquid)
Rb^{88}	0.25	Kr^{85} (4.4 hr)	0.096
Rb^{89}	0.032	Kr^{85} (10.3 yr)	1.5
Sr^{89}	0.036	Kr^{87}	0.19
Mo^{99}	0.029	Kr^{88}	0.024
Te^{101}	0.033	Xe^{133}	24.2
I^{131}	1.6	Xe^{135}	1.5
Te^{132}	2.2	Xe^{138}	0.046
I^{132}	2.1	Corrosion Products	
I^{133}	2.1	Isotopes	Activity, $\mu\text{Ci/ml}$
Te^{134}	0.15	Mn^{56}	0.52
I^{134}	0.31	Co^{60} (5.3 yr)	0.077
I^{135}	0.94	Fe^{59}	0.052
Cs^{137}	0.088	Na^{24}	0.15
Cs^{138}	0.14	Cr^{51}	0.8
Cs^{139}	0.026	Mn^{54}	0.116
Ba^{139}	0.025	Fe^{55}	0.12
		Co^{58}	0.84
		Cu^{64}	0.019

NOTE: Values are design estimates on a 1% defect in fuel rods.

air ejector exhaust. It should be noted, however, that a fuel element failure and a steam generator leak must occur simultaneously if the radioactive gases are to appear in the air ejector exhaust. One such instance occurred at Indian Point and resulted in a gross β , α concentration in the superheater stack of $1.8 \mu\text{Ci}/\text{m}^3$.

Gas stripping columns are used to further remove radioactive gases from coolant water. The stripped gases are incorporated into the gas system used to flush gases from the depressurizer flask tank and to form a gas blanket over liquid storage tanks. At infrequent intervals gas from this system is removed and stored in decay tanks.

Boiling Water Reactors

The Dresden, Big Rock, and Humboldt Bay reactors use the direct boiling cycle as illustrated in Figure B-1 (Sketch B). The radionuclides generated are similar to those generated in a pressurized water reactor. However, the wastes, particularly the gaseous wastes, are significantly different because of the shorter retention of the radionuclides in the coolant system.

In the boiling water cycle, the steam formed from the coolant and containing radioactive gases passes directly to the turbine. The steam condenses, and the noncondensable gases exhaust through the main condenser air ejector to the waste system. Controlled leakage around the turbine seals also carries radioactive gases into the waste system.

The radiogases derive from fission products and activation of gases in

the system. The fission gases consist of the noble gases—krypton and xenon—and may include the halogens—bromine and iodine. These fission gases appear in the coolant because of the fission of "tramp" uranium in the system or failure of the fuel cladding to contain them. The principal activation gas, in terms of total activity present, is usually radionitrogen. It decays with a half-life of a few minutes and, therefore, has no waste disposal significance. The activation of argon gas, comprising about 1 vol% of normal air, creates another radiogas. The argon, which is activated, can be present in air used to ventilate portions of the reactor cavity and in air dissolved in the coolant.

An idea of the relative concentrations and quantities of the waste gases generated by a boiling reactor can be obtained from the Dresden reactor experience which has been well described. For normal operation, activity discharge may be in the order of 100 to 1000 $\mu\text{Ci}/\text{sec}$, consisting primarily of xenon and krypton. If the reactor is operated with maximum tolerable fuel defects, i.e., minor defects in 1.4% of fuel envelopes, stack release activity may reach 0.7 Ci/sec. The principal source of radioactive gaseous waste is the main condenser air ejector. If there are no fuel ruptures, the air ejector exhaust contains a complicated mixture of krypton and xenon fission product gases which presumably from the fission of "tramp" uranium existing on the fuel element surfaces. The estimated release rates are given in Table B-VII. It should be noted also that 35 scfm of gas is discharged by the main

TABLE B-VII. *Estimated Fission Product Gas Release at Dresden*^(a)

<u>Nuclide</u>	<u>Half-Life</u>	<u>Fission Yield</u>	<u>Percent of Activity of Mixture Due to Each Nuclide After 20 min Holdup</u>
Kr ⁸⁹	3.2 min	4.6	1.8%
Xe ¹³⁷	3.8 min	5.9	4.2
Xe ¹³⁵ min	15 min	1.8	10
Xe ¹³⁸	17 min	5.5	31.4 *
Kr ⁸⁷	78 man min	2.7	13.5 *
Kr ⁸³ min	1.86 hr	0.48	2.1
Ki ⁸⁸	2.8 hr	3.7	14.2 *
Kr ⁸⁵ min	4.4 hr	1.5	4.7 *
Xe ¹³⁵	9.2 hr	6.2	14.0 *
Xe ¹³³ min	2.3 day	0.16	0.1
Xe ¹³³	5.27 day	6.5	3.9 *
Xe ¹³¹ min	12.0 day	0.03	-
Kr ⁸⁵	10.4 yr	0.3	-
			100%

(a) From "Report of Gaseous Effluent Monitoring and Control Practices for Dresden Nuclear Power Station," Docket 50-10, USAEC Public Document File, August 1, 1961.

* Six radionuclides account for 80% of the total activity.

condenser air ejector; and that this gas requires 15 to 20 min to travel through 130 ft of 30 in. pipe to where it is filtered, mixed with 44,000 scfm of ventilation air, and discharged out the stack. A maximum of 200 scfm of air is contained in the steam leaking around the turbine gland seals. The gases remaining after the steam is condensed are passed through 130 ft of 24 in. diam pipe which provides a two minute delay to the stack. The two gas streams, i.e., (1) the diluted

exhaust from the condenser air ejector, and (2) the gases leaking around the turbine gland seals, discharge to the stack; there they mix with 44,000 scfm of ventilation air from the turbine building.

Sodium Cooled Reactors

The Hallam and Fermi reactors employ used sodium as a coolant in a system as illustrated in Figure B-1 (Sketch C). The corrosion rates of reactor materials in contact with the hot sodium

(800 to 1000 °F) is very low. Consequently, the concentration of activation products in the coolant system is also very low, and the principal sources of radioactivity are activation of the sodium itself and from impurities in the sodium. Sodium-24,

sodium-23, rubidium-86, and antimony-124 are formed but these are of no environmental significance as far as out-plant because they are effectively contained in the coolant system until removed by a cold trap.

APPENDIX C

A SEQUENTIAL SAMPLING PROCEDURE

A sequential sampling procedure as referred to in the text is the plotting of cumulative totals after successive observations as illustrated essentially by Figure C-1. Note that the chart on which the data are plotted contains two boundary lines. When the cumulative total crosses one of the boundary lines, conditions are said to be "approved" or "not approved".

This sequential sampling procedure depends upon the effluent concentrations or rate of release being normally distributed; the procedure breaks down if the distribution is not normal. However, if the distribution is lognormal, a modified procedure can be used which involves plotting the cumulative product of the data against the number of results. The corresponding limit lines then become curves. A few years of operation may show that the assumption of a normal distribution is valid. Until sufficient data are available from which to reliably determine the distribution of radioactive materials within the effluents, (if, indeed, that is ever possible) it is necessary to sample frequently enough to include all changes in concentration or rate of release that could be significant. The number of people and amount of laboratory equipment required to adequately sample a flowing stream, whose properties change rather rapidly, make such an effort impractical to continue

indefinitely on an operational basis. A continuous onstream monitor is the obvious alternative and if an acceptable continuous monitor cannot be designed, then a complete sample survey for a period of time long enough to reliably establish operational parameters is required.

The position of the boundary lines depends on the conditions specified for the test. If it is assumed that the sample values are distributed normally, and that there should be a probability α (say 0.001) of disapproving the discharge of a waste if $m = \mu - 2\sigma$ where m is the average concentration and μ is the MPC for approving the discharge when $m = \mu + 3\sigma$, then the limiting lines (τ_e and τ_μ) would be given by:

$$\tau_e = n\mu - \frac{\sigma}{4} \ln\left(\frac{1-\alpha}{\alpha}\right)$$

$$\tau_\mu = \mu + \frac{\sigma}{4} \ln\left(\frac{1-\alpha}{\alpha}\right).$$

τ_μ and τ_e are the upper and lower boundaries, and n is the number of samples. For purposes of illustration of this method, certain effluent data from the PRTR are plotted in Figure C-1. The arsenic-76 concentration before dilution in the liquid waste is shown by the solid line. The boundaries are shown as dashed lines. The MPC_w for As^{76} is listed (in 10 CFR, Part 20,

Appendix B, Table B-II) as 2×10^{-5} $\mu\text{Ci}/\text{ml}$. By the end of the fifth sampling period, the waste is demonstrably "acceptable" for discharge. It is necessary to assume that the condition of the waste in question remains constant over the time interval selected between samples for this approach to be valid or, alternatively, use of an auxiliary continuously monitoring device

capable of detecting abnormally high concentrations. Such "emergency" monitoring devices are commonly used at the present time. A study of monitoring data available from operating power reactor plants would be an important prerequisite to the development of such a sequential sampling system in the atomic power industry.

FIGURE C-1. Sequential Test of PRTR Data

APPENDIX D

FACTORS AFFECTING THE DOSE DELIVERED
BY RADIOIODINE AND THE RADIOACTIVE
NOBLE GASES

Iodine

At least 24 isotopes of iodine occur as fission products or daughters of fission products. These have mass numbers ranging from 127 to 140 and all are radioactive except I^{127} . Only the isotopes I^{131} through I^{135} have any environmental contamination significance following the release of fission products.

The relative activity of I^{131} through I^{135} as a function of time is shown in Figure D-1. It can be seen that I^{131} contributes an increasing portion of the total activity starting at several hours after fission. Except for exposures involving the inhalation of radioiodines that are only a few hours old, the dominant exposure is contributed by I^{131} .

The marked ability of the thyroid organ to selectively concentrate and retain iodine plus the fact that it is a relatively small organ makes it the "critical organ" when the inhalation or ingestion of radioiodine is involved.

Iodine released to the atmosphere in gaseous and particulate form concentrates in the environmental pathway which leads from release to deposition on grass to the milk of cows that eat the grass. The thyroids of persons who drink the milk receive doses that are many times greater than the dose

from inhalation of the released iodine. Soldat (1963) reports a factor of 400 times for the potential dose received by a small child (with a 2 g thyroid) via milk as compared to inhalation. He estimates the ratio of these doses in an adult (20 g thyroid) to be about 40 to 1. An estimated dose ratio of 80 to 1 is reported in IDO-12035 (1964) which describes the Controlled Environmental Radioiodine Tests at the National Reactor Testing Station.* [It should be noted the use of ingestion to inhalation dose ratios assumes that certain parameters such as breathing rate, uptake fraction and daily milk intake are the same].

Facilities that release radioiodine must apply permissible release rates which take into account the environmental reconcentration of the iodine. The relative dose factor described in the previous paragraph can be used conveniently for this purpose. For example, if the dose from milk is 400 times the dose from air (inhalation) the MPC's can be multiplied by 1/400 to obtain a concentration in air which should not lead to above permissible thyroid doses from drinking milk. Factors of this type were first developed

* More recent results of the CERT study are reported in IDO-12047.

FIGURE D-1. Calculated Activity of Noble Gases, Krypton + Xenon in PRTR Fuel After 10,000 MWd/T - 619 Day Irradiation

at Hanford and are in use at several power reactors. The Bonus reactor, for example, operates with a permissible stack discharge limit which is 1/1200 the limit that would apply if inhalation of radioiodine and not ingestion via milk were the principal exposure pathway. A factor of 1/700 is applied at the Elk River and Pathfinder sites.

Noble Gases

The occurrence and release of the inert fission and activation gases play an important part in the establishment of permissible stack release limits and the monitoring of gaseous effluents. The actual release of these gases from power reactors has created no radiation protection problems. Indeed, during the years 1960 - 1963 only 0.13% of the permissible amount of gaseous wastes was actually released by all power reactor plants. The maximum percentage release by any single plant during any one of those years was 0.33%. Measurable off-site doses from noble gases were observed during the end of the last operating cycle at Humboldt Bay Unit No. 3. Stack release rates during this period increased to the noble gas release limit of 50,000 $\mu\text{Ci/sec}$ because of defective fuel cladding. The maximum off-site dose was 1/3 the annual 500 mrem permissible dose.

Several isotopes of krypton and xenon gas occur as fission products.

Figure D-2 illustrates the relative activity levels with time of these isotopes after a specific irradiation time in the PRTR reactor. Other relatively inert gases such as argon-38, nitrogen-16 and nitrogen-13 are formed by activation of constituents of air in the reactor neutron flux. The amounts of the fission gases formed can be accurately predicted because they are a function of the reactor power level. Accurate prediction of the amount of activation gases formed requires accurate knowledge of the quantity of air irradiated and the duration of the irradiation.

The particular mixture of the radioactive gases appearing in the stack can differ considerably among reactor plants because it is a function of many variables. One of the most important factors is the time that elapses between generation of the gas and its appearance in the stack. After one day the only krypton isotope of any consequence is Kr^{85} while Xe^{133} contributes nearly all of the activity from the xenons.

Because the inert gases are not selectively metabolized or concentrated in particular organs by the body, the radiation dose of interest is that delivered by a cloud of the gas. The published MPC's for the inert radioactive gases are calculated for immersion in a semispherical infinite cloud.

FIGURE D-2. Calculated Activity of
Radioiodine Isotopes in PRTR Fuel
Following 10,000 MWd/T - 619 Day
Irradiation

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