GUIDANCE FOR AIR SAMPLING AT NUCLEAR FACILITIES

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ABSTRACT

The principal uses of air sampling at nuclear facilities are to monitor general levels of radioactive air contamination, identify sources of air contamination and evaluate the effectiveness of contaminant control equipment, determine exposures of individual workers, and provide automatic warning of hazardous concentrations of radioactivity. These applications of air sampling are discussed with respect to standards of occupational exposure, instrumentation, sample analysis, sampling protocol, and statistical treatment of concentration data. Emphasis is given to the influence of spacial and temporal variations of radionuclide concentration on the location, duration, and frequency of air sampling.

TABLE OF CONTENTS

	<u>Page</u>
1. Introduction	1
2. Standards for Occupational Exposure.	3
2.1 Sources	3
2.2 Modes of Exposure	3
2.3 Application	4
3. Objectives of Air Sampling.	8
4. Air Sampling Instrumentation	.10
4.1 Selection.	.11
4.1.1 Mode of Measurement	.11
4.1.1.1 Special cases	.11
A. Beta- and gamma-emitting noble gases	.11
B. Tritium.	.12
C. Radon	.12
4.1.2 Sensitivity.	.13
4.1.3 Convenience.	.14
4.1.4 Reliability.	.14
4.1.5 Accuracy.	.14
4.1.6 Power Requirements	.15
4.2 Collectors and Detectors.	.15
4.2.1 Direct Reading Air Samplers	.15
4.2.2 Accumulative Air Samplers	.16
4.2.2.1 Filter media	.16
4.2.2.2 Filter holders.	.18
4.2.2.3 Other collection methods	.19
4.2.2.4 Automatic instruments	.20
4.2.3 Collectors for Measurements of Particle Size	.20
4.3 Pumps	.22

TABLE OF CONTENTS (Cont'd)

	<u>Page</u>
4.3.1 Low Volume Pumps	.24
4.3.2 High Volume Pumps.	.24
4.3.3 Personal Sampler Pumps	.25
4.4 Flowmeters	.25
4.4.1 The Rotameter	.25
4.4.2 Fixed Orifice Meters.	.25
4.4.3 The Critical Orifice.	.26
4.4.4 Flowmeter Calibration	.26
4.5 Precautions	.28
5. Sample Analysis	.29
5.1 Types of Measurement	.29
5.2 Requirements for Counting Instruments	.29
5.3 Types of Instrument	.30
5.3.1 Ionization Chambers	.30
5.3.2 Proportional Counters	.31
5.3.3 Geiger Counters	.31
5.3.4 Scintillation Counters	.31
5.3.5 Semiconductor Detectors.	.31
5.4 Application of Instrumentation to Sample Analysis	.32
5.4.1 Alpha Counting.	.32
5.4.2 Alpha Spectrometry	.32
5.4.3 Beta Counting	.32
5.4.4 Gamma Counting.	.32
5.4.5 Gamma Spectrometry	.32
5.5 Precautions	.33

TABLE OF CONTENTS (Cont'd)

	<u>Page</u>
6. Protocol	.34
6.1 Characteristics of Work Atmospheres	.34
6.2 Types of Sample.	.35
6.2.1 General Air (GA) Samples	.35
6.2.2 Breathing Zone (BZ) Samples	.36
6.2.3 Personal Air (PA) Samplers.	.36
6.2.4 Process Samples	.36
6.2.5 Precautions.	.37
6.3 Monitoring General Air Concentrations	.37
6.3.1 Placement of Samplers	.38
6.3.2 Sampling Frequency	.39
6.3.3 Types of Sampling Instruments.	.39
6.3.4 Data Interpretation	.39
6.4 Monitoring Employees' Exposures	.40
6.4.1 Monitoring Exposures with Personal Samplers.	.41
6.4.2 Monitoring Exposures with Breathing Zone (BZ) and General Air (GA) Samples.	.43
6.4.3 Monitoring Exposures with General Air Samples	.46
6.4.4 Particle Size Measurements.	.47
6.5 Emergency Monitoring	.49
7. Statistical Methods	.53
7.1 Measurement Precision.	.53
7.2 Sample Averaging	.55
7.3 Comparing Average Concentrations	.56
7.4 Log-Normal Distributions.	.60
7.5 Determination of Number of Samples	.61
7.6 Precision of a Time-Weighted Exposure	.64
7.7 Effect of Sampling Period on Sample Coefficient of Variation.	.64
References	.66

1. INTRODUCTION

Most nuclear processes, from research involving minute quantities of radioisotopes to the several phases of the atomic power cycle, give rise to radioactive air contaminants that can be hazardous to workers if not adequately controlled. Adequate control is achieved by various safeguards, mainly containment and ventilation, that maintain occupational exposures at safe levels in accordance with pertinent standards. Effectiveness of control, i.e., conformance with standards of exposures, must be verified by monitoring, which, for the majority of radionuclides, is done most reliably by air sampling.

There are two aspects of air sampling that must receive equal consideration in a properly executed monitoring program. The first is the method by which a sample is collected and analyzed to yield an accurate measurement of an airborne radionuclide. The importance of this is readily apparent. The second aspect is the protocol covering sampling location, duration and frequency by which the safety of the occupational environment is reliably assessed. The critical nature of this point may be less obvious. By a single air sample, a measurement is obtained of radionuclide concentration only at the particular location and time of collection. This rarely constitutes adequate information because concentrations vary tremendously with time and location and are often affected by local work activities. Repeated air samples, distributed by time and location, are more indicative of general conditions and, hence, the safety of the occupational environment. By additionally taking account of work routines, and, in some instances, by sampling at specific manual operations, the exposures of individual employees to airborne radionuclides can be determined, a practice that is recommended when air concentrations approach permissible levels. Thus, protocol of an air sampling program must be determined with great care and must be adapted to the particular size and nature of each nuclear facility.

The extreme variability in radionuclide concentrations common to most facilities complicates the interpretation of air sampling data. Indeed, variability with time and location undoubtedly constitutes the greatest difficulty in deciding on frequency and distribution for a sampling program and there is scant quantitative information available for guidance. Therefore, initial protocol must be more or less arbitrary but as pertinent data accumulate it can be adjusted to yield acceptable accuracy. Properly adapted to local conditions, air sampling can provide a very reliable

indication of the quality of the occupational environment, and if warranted, can provide reliable values of individual exposure.

The principal uses of air sampling are to monitor general levels of radioactive air contamination, identify sources of air contamination and evaluate the effectiveness of specific contaminant control equipment, determine exposures of individual workers, and provide automatic warning of hazardous concentrations of radioactivity. The first three uses tend to overlap in many ways with respect to procedures and equipment and are reflected to some extent in most air sampling programs. The warning function involves special instrumentation and is restricted to facilities having a reasonable risk of serious, acute hazard.

The need for air sampling is governed by the likelihood that significant concentrations of radioactivity (relative to pertinent standards) may occur in the work environment. Some airborne contamination is apt to occur wherever unencapsulated radioactive materials are handled but the magnitude of air concentrations is strongly dependent on the type of process and on the quantity and form of the radionuclide. No reliable, general criteria exist for predicting air concentrations; precedent and judgement are usually invoked in deciding on air sampling needs. In doubtful cases, exploratory air sampling is conducted to determine if air contamination is a significant hazard.

Air sampling often is used in combination with other monitoring techniques to increase confidence in environmental safety. Bioassay (including in-vivo counting), surface contamination monitoring, and external radiation monitoring may be used in varying degree in conjunction with air sampling. Relative emphasis among the several techniques depends on ease of measurement and biological behavior of the radionuclide.

This guide stresses the principles and practices of air sampling. Instrumentation and analysis are covered to the extent necessary for rational selection and application.

2. STANDARDS FOR OCCUPATIONAL EXPOSURE

Biological injury from exposure to an airborne radionuclide depends directly on the amounts of the radionuclide accumulated in critical organs of the body. These are the basic standards, but to facilitate control of the working environment, standard-setting authorities calculate air concentrations at which continuous exposure does not lead to excessive body burdens. These air concentrations, designated "maximum permissible concentrations" (MPCs) are the standards used for control and monitoring at nuclear facilities. All objectives of air sampling programs relate to these concentration standards.

2.1 Sources

The International Commission on Radiological Protection (ICRP) is a prestigious, non-governmental organization with world-wide scientific representation from which nearly all recognized standards of radiation exposure are derived. Actually, ICRP only recommends standards but, with few exceptions, the recommendations are adopted directly by standards-setting agencies. In the United States, the National Council on Radiation Protection and Measurements (NCRP) also recommends radiation protection standards and although NCRP and ICRP are independent, their respective recommendations are virtually identical because of close collaboration between the two organizations.

Two U. S. federal agencies have statutory authority to set radiation standards the Nuclear Regulatory Commission (NRC) and the Environmental Protection Agency (EPA). Most uses of radioactive materials in the United States must be in accordance with standards set by these two agencies. The Energy Research and Development Administration (ERDA) sets standards for its own facilities but these are closely related to the NRC standards. The NRC promulgates standards of occupational exposure for its licensees in the Code of Federal Regulations Title 10 Part 20. Part 20 radiation standards also apply to licensed radioisotope users operating in states that exercise control by agreement with the NRC. EPA sets standards of exposure for the general public and for radiation facilities outside the jurisdiction of NRC and ERDA.

2.2 Modes of Exposure

Standards are set with regard to the mode of exposure having the most likely risk of biological damage. For the majority of

radionuclides, the critical mode of exposure to a worker in a radioactive atmosphere is by inhalation, that is, the internal radiation dose from radionuclides that enter through the respiratory system represents the dominant risk of injury. For certain beta-emitting and gamma-emitting noble gases, the external, whole-body radiation dose from immersion in the gas exceeds the dose from gas within the body and therefore constitutes the critical mode of exposure. In the case of one radioactive substance, tritiated water vapor, skin absorption and inhalation are exposure modes of comparable importance.

For the so-called immersion gases, e.g., the radioactive argons, kryptons, and xenons, atmospheric monitoring is best achieved by measurement of the external radiation field using suitably calibrated survey instruments but for all other radionuclides, atmospheric concentrations are necessarily measured by air sampling techniques.

2.3 Application

Certain aspects of MPC derivations should be taken into account in conducting an air sampling program.

First, MPCs are set so that the rate of accumulation of a radionuclide in the body organ of interest will not result in an annual dose rate exceeding the basic standard after an equilibrium quantity of the radionuclide exists in the organ. Therefore, the MPCs are average limits and can be exceeded temporarily without violation if there are compensating periods of exposure at lower concentrations. Generally, it is advisable to avoid this kind of situation by maintaining radionuclide concentrations well below MPCs, but occasional excursions above the MPC are not always preventable.

Second, MPC calculations are inexact. The actual amount of inhaled radionuclide that accumulates in a particular organ and the resultant radiation dose to that organ depend on a host of factors including the physical and chemical properties of the air contaminant, the respiration pattern of the subject, rates of biological transfer, and organ size, all of which are highly variable and some of which are not known accurately. In any case, these variable parameters are standardized in order to promulgate a single MPC for each radionuclide that will be valid in the broadest possible circumstances. The significance of this procedure in deriving the MPC is twofold: 1) MPCs are imprecise. This doesn't compromise safety because the calculations are con-

servative. But it does remove any inferred cloak of sanctity that might attach to the values. 2) If warranted, the precision can be improved in particular circumstances by replacing the standardized factors with measured data. (Licensees must obtain approval from appropriate authorities for any departure from standard values.) Ordinarily, this kind of substitution is only feasible with respect to properties of the air contaminant because biological parameters are difficult to measure in individual subjects. More specifically, the solubility and chemical form and, in the case of an aerosol, the particle size, all of which are often amenable to measurement, may be used to adjust an MPC or more usually, to obtain improved estimates of organ burdens or doses in particular exposures.^(1,2) Although procedures have not yet been standardized, guidance on the use of particle size measurement is given in section 6.4.4.

Maximum permissible concentrations in air for well over 200 radionuclides are listed by ICRP,^(1,3) NCRP,⁽⁴⁾ and in CFR 10, Part 20,⁽⁵⁾ representing most of the radionuclides normally encountered. MPCs for unlisted radionuclides can be derived according to methods given by ICRP⁽¹⁾ and NCRP.⁽⁵⁾ MPCs are also given for unknown mixtures of radionuclides.

If two or more radionuclides are present as artificial air contaminants, it may be necessary to derive a composite MPC. For this purpose, the mixture rule is invoked by which the respective MPCs are weighted by the relative concentrations of the corresponding radionuclides. The following formula is used:

$$\frac{C_A}{MPC_A} + \frac{C_B}{MPC_B} + \dots + \frac{C_N}{MPC_N} \leq 1$$

where C_A , C_B , C_N are the concentrations of radionuclides A, B, and N.

MPCs are expressed in units of microcuries per cubic centimeter of air ($\mu\text{Ci}/\text{cm}^3$). They are based on an occupational exposure of 40 hours per week, 50 weeks per year. For shorter or longer work schedules, the values may be increased or decreased proportionately.

A direct interpretation of the MPC value is that no individual should be exposed to an average concentration in excess of the value. However, different averaging periods are specified by standards-setting organizations. In CFR 10, Part 20, the NRC stipulates 40 hours in any period of seven consecutive days. By

contrast, ICRP and NCRP recommend a quarter of a year and, furthermore, permit as much as a half-year's exposure in one quarter, provided that the average exposure for the full year does not exceed the MPC. The ICRP-NCRP recommendation permits greater flexibility in the management of exposure but the NRC rule more closely reflects the cyclic nature of most routine work. In either case, fairly detailed records of time-weighted exposures may be necessary in order to prevent exposures in excess of permissible. These can be avoided if all concentrations are maintained below MPC values so that conditions conducive to excessive exposures do not exist.

The degree of air sampling required is strongly related to the likelihood that occupational exposures might exceed MPC values which, in turn, depends on the type of facility and the radionuclides in process. In marginal cases, at least some exploratory sampling is necessary to determine whether airborne radionuclides occur in significant concentrations. Unless no air contamination is detected, it is prudent to recheck periodically, the frequency of rechecks depending on the probability of abnormal air concentrations but as a minimum, quarterly.

ICRP⁽⁶⁾ recommends special monitoring for employees who may be exposed to more than 3/10 of the annual maximum permissible dose. This involves measurements in sufficient detail to determine individual exposures. Where exposures are expected to be less than 3/10 of the annual maximum permissible dose, it is deemed adequate to monitor the working environment in order to verify that this criterion is met.

CFR 10. Part 20 specifies personal monitoring equipment for employees likely to be exposed to external radiation greater than 25% of the quarterly permissible dose but is mute regarding any need for special monitoring of airborne radionuclides. On the other hand, it does require the licensee to implement surveys that enable him to comply with regulations and this implies that more detailed air sampling is required as exposures approach MPCs.

Despite the fundamental effects of particle size and solubility on biological hazard, these properties of airborne radionuclides are seldom measured in routine monitoring programs because in theory, MPCs are compensated for the effects, enabling monitoring to be based on measurement of total air concentration. Moreover, adjustments of the MPC or of dose for different particle sizes or solubilities are rather complex and are not encouraged by regulatory agencies.

Compensation in the derivation of MPCs by ICRP is done by:

1. assuming a fractional lung deposition of the inhaled radionuclide compatible with a particle of average size, and
2. calculating separate values for soluble and insoluble forms of each radionuclide.

Although somewhat arbitrary in its selection (by ICRP), the fractional deposition used for the lung is approximately correct over a range of activity median diameters from about 0.3 to 3.0 μm . At sizes smaller than 0.3 μm , lung deposition tends to increase above the assumed fraction while at sizes larger than 3.0 μm it diminishes. Therefore, the neglect of particle size in monitoring occupational exposure generally does not result in increased risk except for very small particles.

Deciding on solubility in selecting the MPC for the particular radionuclides at a nuclear facility can be of critical importance because differences between the MPC values for the "soluble" and "insoluble" forms are substantial in many cases. But neither standards organizations nor regulatory agencies define solubility and the decision is left to local judgement. Useful guidance is given by the ICRP Task Group on Lung Dynamics but this has not yet been officially adopted by ICRP nor has it been recognized in CFR 10, Part 20. Consequently, by custom, selection is based on handbook solubilities of the bulk compounds. For compounds of intermediate solubility, the choice is difficult and usually the lower of the two MPC values is chosen in the interest of safety

3. OBJECTIVES OF AIR SAMPLING

The basic reason for air sampling is to ascertain the safety of the atmospheric environment and to provide information on which to base improvement, if needed. However, in achieving its fundamental purpose, air sampling may be tailored to fulfill various specific objectives.

In the most common situation, air sampling provides continuing assurance that the atmospheric environment is inherently safe for routine occupational pursuits. This applies to the desirable circumstances wherein radionuclide concentrations rarely if ever exceed the MPC and for the most part remain considerably below the MPC. This condition would be consistent with the ICRP exposure category in which resulting doses are most unlikely to exceed 3/10 of the annual maximum permissible dose. Sampling, therefore, is focused on working areas with special attention to critical locations where abnormal concentrations are most likely to occur. Demonstration that concentrations in these locations are below the MPC constitutes reassurance that all work areas are satisfactory. Samples collected for this purpose are designated "general air" samples.

In less frequent circumstances, it is found that not all concentrations can be maintained below the MPC and there is the likelihood that average exposures of at least some employees will approach or exceed the MPC. This situation, which may exist only in certain well-delineated portions of a facility or may be general, corresponds to ICRP exposure conditions wherein the resulting doses might exceed 3/10 of the annual maximum permissible dose. Accordingly, sampling objectives must be expanded to include monitoring individual exposures. Thus, in addition to general air sampling, provision is made to measure the specific, often high concentrations which individual workers encounter in the course of the work day. For this purpose, "breathing zone" samples are collected by means of portable air samplers or special personal samplers worn by the workers. Monitoring is done in a manner that enables average exposure to be determined for each worker for a designated time interval such as a week or a month. An obvious corollary to this is a procedure whereby excessive exposures are identified promptly and prevented from recurring.

An important application of air sampling in all nuclear facilities is to determine the effectiveness of contaminant control equipment such as enclosures, hoods, and glove boxes. Actually, all air samples serve this purpose to some degree by indicating

the general state of effectiveness and occasionally pinpointing sources of contamination that arise through malfunction, deterioration, or human error. However, additional special sampling may be performed to evaluate the control provided by particular items of equipment that are newly installed or otherwise suspected to be sources of air contamination. This technique should be applied during the initial operation of a new facility to be sure that control measures are functioning as intended. Samples collected for control evaluation are apt to differ from conventional general air samples in that they may be taken in locations that are inaccessible to employees. These are designated "process samples" and should not be used for exposure evaluation.

Air monitors are used as alarms in facilities having the potential for an acutely hazardous release of radionuclides that wouldn't be detected by other means. The need is generally restricted to facilities that process a substantial quantity of a radionuclide having a high specific activity and a low MPC. The alarm provides warning to evacuate or to don protective equipment and to initiate other emergency procedures. It also indicates the magnitude of the air concentration and may be arranged to activate other monitoring devices specifically intended to register accidental releases. Resultant information can be helpful in estimating dose to affected employees.

All of the foregoing air sampling functions can be fulfilled by measuring the gross concentration of individual radionuclides, the practice followed in the vast majority of monitoring programs but for excessive exposures, it may be desirable to improve the quality of exposure information by means of supplementary techniques. Foremost among these are measurements of the size of particulate radionuclides which enable more accurate assessments of respiratory deposition to be made. Particle size classifying devices are sometimes incorporated in routine sampling programs, while in others their use is limited to special investigations.

4. AIR SAMPLING INSTRUMENTATION

Air sampling instruments exist in a wide range of styles and capabilities, the characteristics of individual types usually being suited to prescribed applications. They range in size from conveniently portable units to large, permanent installations and in air capacity from a few liters per minute to more than a cubic meter per minute. Ordinarily air samplers are procured from commercial sources as complete units but in some of the ensuing discussions the basic components will be treated separately for clarity.

The basic components of an air sampler are a collector (or detector), an air flowmeter, a valve to adjust the flow rate, and an air mover and they are usually connected in that order. In particulate samplers, the collector is always placed ahead of the other components (with respect to the direction of air flow) to avoid particle losses in connections, valves, etc., prior to collection. This is considered more important than placing the air flowmeter first, which would avoid the reduction in air density at the flowmeter imposed by the collector, because a correction can readily be made in the flowmeter calibration to account for air density whereas there is no feasible method to correct for sample loss. However, the valve for air flow regulation is positioned after the flowmeter to avoid any additional alteration of the air density at the flowmeter.

Both direct-reading and accumulative air monitors are used for measuring radionuclide concentrations although direct-reading instruments are mainly limited to measurements of radioactive gases. Accumulative sampling, predominantly by collection on filter paper, is the most widely employed mode of air monitoring in the nuclear industry. In general, direct-reading instruments provide the immediacy that is useful for engineering purposes and is vital in emergencies but they have finite, sometimes marginal, sensitivities. Results of accumulative sampling are delayed by the time interval required for analysis but virtually any sensitivity can be achieved by collecting appropriate sample volumes. The inherent delay is not a handicap for routine monitoring but sharply limits the usefulness of accumulative sampling for emergencies. This limitation prompted the development of automated filter samplers which give continuous or periodic readings of accumulated radioactivity. These instruments comprise a third "class" between the direct-reading and accumulative types. In addition, there are special types of (accumulative) collecting devices for measuring the physical and chemical properties of

particulate radionuclides.

4.1 Selection

Key factors in selecting air sampling instruments are: appropriate mode of measurement, sensitivity, convenience (size, weight, ease of operation), reliability, accuracy, and power requirements.

4.1.1 Mode of Measurement. Direct reading instruments are usually intended for monitoring just one radionuclide or a class of related radionuclides and this information is given in specification circulars. However, an inquiry to the manufacturer is advisable if local conditions such as high humidity, corrosive gases, abnormal external radiation fields, or other airborne radionuclides might affect instrument performance.

For most accumulative sampling, the mode of measurement after sample collection is selected by matching of analytical method to the collected radionuclide. Usually radiometric analysis is used with or without some preparation of the sample. Collection on a filter paper followed by direct alpha, beta or gamma counting is the simplest and most common procedure. More elaborate analysis including chemical separation and/or energy spectrometry is used for mixtures of radionuclides. Methods of sample analysis are described in detail in the HASL Procedures Manual (HASL-300, Health and Safety Laboratory, USERDA, New York, NY) and elsewhere.

For some radionuclides, special collection media must be used. For example, radioactive iodine may exist in several chemical forms necessitating an array of several different sampling media to assure quantitative collection. Radioactive gases not susceptible to direct reading methods are sampled by adsorption or collection of a fixed volume in containers.

4.1.1.1 Special cases. Several gases warrant brief comment because of special characteristics.

A. Beta- and gamma-emitting noble gases. The dominant mode of exposure to these gases in ambient air is external radiation and although the MPCs, like those of all other radionuclides, are expressed in $\mu\text{Ci}/\text{cm}^3$, they are calculated to limit external, whole-body radiation exposures to 5 rem/year. Therefore, the most direct means of monitoring is a conventional radiation survey meter or radiation dosimeter having an energy response appropriate to the particular type of emission. The applicable radiation guide corresponding to the MPCs is 2.5 mrem/hr.

Circumstances may require measurements of actual gas concentrations, as well. For example, it might be necessary to distinguish between external radiation from a gas in ambient air and from another source in the same area in order to select an appropriate method of control. Several kinds of instruments are available for this, the most useful being flow-through gas monitors utilizing Geiger-Muller or scintillation detectors (4.2.1). These must be calibrated for the specific radionuclide and require shielding if they are to be operated directly in the environment containing the gas or in the presence of any other significant external radiation. A typical instrument of this type has a sensitivity of 10^{-6} $\mu\text{Ci}/\text{cm}^3$ for krypton-85.

B. Tritium. Tritium may exist in air as either hydrogen gas, as an organic compound or as a component of water vapor, the latter having much more restrictive MPC because it is completely absorbed by body fluids during inhalation. Tritium is difficult to measure because of its very weak beta energy of 18.6 keV. Flow-through ionization chambers, popularly known as Kanne chambers (4.2.1), are the most common instruments used for air monitoring. These instruments are subject to interference from external radiation, other radioactive gases, and aerosols. The interferences are counteracted with partial effectiveness by a variety of methods including the attachment of sealed ionization chambers of identical size to provide a compensating signal responsive only to external gamma radiation and an electrostatic precipitator or filter to remove atmospheric particulates from the entering airstream. The sensitivity of a typical instrument is 10^{-6} $\mu\text{Ci}/\text{cm}^3$.

Where greater sensitivity is required or interfering air contaminants are particularly troublesome, tritiated water vapor can be sampled by adsorption on silica gel or by a water bubbler and then analyzed in a liquid scintillation counter (4.2.2.3)

C. Radon. The hazard caused by radon, an alpha-emitting noble gas, is the inevitable presence of its short-lived decay products, known as radon daughters. Therefore, the most direct assessment of hazard consists of measuring the concentration of its daughters but often it is preferable to measure rado itself for control purposes. Several techniques are available of which the alpha scintillation flask and the two-filter tube are in most common use. An air sample is collected and sealed in the scintillation flask which is made of glass or clear plastic, has a volume of 100 to 500 ml, and is coated internally with zinc sulfide phosphor. For counting, the flask's flat, transparent base is placed on a photocathode tube which detects scintillations

produced at the flask's inner walls. In the case of the two-filter tube, air is drawn through the tube which is equipped with high efficiency filters at both its inlet and exit. In transit through the tube, a small fraction of radon contained in the sample air decays to polonium-218 atoms which are solid and thereby are collected on the exit filter. After a suitable collection period of 5 to 10 minutes, the exit filter is removed and promptly alpha-counted. Radon concentration is directly proportional to the alpha activity on the filter paper for a given tube, flow rate, and sampling period.

4.1.2 Sensitivity. Required sensitivity varies with circumstances but it is generally desirable to be able to measure concentrations at least to 1/10 MPC. There are obvious exceptions. For example, a sensitivity of 10 MPC or even 1000 MPC might be adequate for an alarm.

The sensitivity of direct reading instruments is fixed and can be found on specification circulars. However, definitions of sensitivity vary and the manufacturer should be asked about the basis for the listed values if this information is not given. A typical definition of sensitivity is the lowest concentration that can be measured with a precision of $\pm 50\%$. Sometimes it is expressed as a multiple of the background reading. Sensitivity may be affected by local environmental conditions, especially external radiation.

The minimum radionuclide concentration that can be measured with an accumulative sampler depends on sample flow rate, sampling period, and analytic sensitivity:

$$C = \frac{q}{TF}$$

where

C = detectable concentration $\mu\text{Ci}/\text{cm}^3$
q = minimal detectable quantity of radionuclide, μCi
T = sampling period, minutes
F = sample flow rate, cm^3/min

Conceptually, any degree of sensitivity can be attained by increasing the sample volume, TF, indefinitely.* This is subject to practical constraints but available instrumentation permits considerable flexibility in flow rate and sampling period. Usually, the decision entails a compromise that enables a small fraction of the MPC, say 1/10, to be measured in a brief time period (no more than 60 minutes for portable equipment, up to 8 hours or possibly 40 hours for fixed samplers), using equipment of convenient size and reasonable cost.

The sensitivity of an alarm monitor is more complicated and may be variable, being dependent on the concentration of natural radioactivity (radon daughters and thoron daughters) in the atmospheric environment. This is another case where characteristics of a particular instrument must be obtained from the manufacturer.

4.1.3 Convenience. Size and weight are important factors of convenience for portable air sampling instruments and are critical in the case of personal samplers. Ease of operation warrants consideration with special attention to the method of changing the collection medium, reading and adjusting air flow and any necessary routine maintenance (oiling, cleaning). Additional factors are battery life (for portable instruments), noise of operation, and temperature rise (especially important for hand-held instruments).

4.1.4 Reliability. Trouble-free operation is mandatory in monitoring equipment. Other users constitute the best source of information on this point. Parts wear, calibration drift, and development of air leakage are common difficulties.

4.1.5 Accuracy. The accuracy of a method for sampling radioactive gas is most reliably measured by sampling from a standard concentration. Manufacturers of direct reading instruments may determine accuracy by this means, or they may rely on indirect calibrating procedures such as comparison with another method or instrument. Direct-reading instruments should be tested at several different concentrations to check for nonlinearity of the scale reading which can be a source of inaccuracy. Five percent deviation is a reasonable limit for nonlinearity.

*An exception is sampling radionuclides with short lives relative to the sampling period ($T_{1/2} < 10T$) wherein accumulation of activity is non-linear with time. In this case, maximum sensitivity is achieved at a sampling period of about six half-lives.

There is no direct method of determining the absolute accuracy of particulate samplers because of the lack of a method of generating a standard concentration of particulate radionuclides. The approach to accuracy therefore must depend on the elimination of anticipated sources of error. Analytic methods are critical in this regard but flowmeter calibration and the absence of air leaks are important. Precautions are discussed in 4.2.2.2, 4.4.4, and 4.5.

4.1.6 Power Requirements. This is usually a minor consideration except for battery-powered equipment, especially personal samplers. Size, weight, charging characteristics, and battery life are then consequential factors.

4.2 Collectors and Detectors

4.2.1 Direct Reading Air Samplers, which provide continuous, instantaneous measurement of air concentration, are available only for radioactive gases. Typically, sample air is drawn continually through a sensitive chamber where radioactivity is measured by ionization current, gas proportional counting, scintillation counting, or a Geiger-Muller detector. The most common direct-reading samplers are for tritium vapor, using an ionization chamber (Kanne chamber), and for radioactive krypton, using a GM tube. Both types have been adapted to other radioactive gases in special applications. In addition, direct-reading samplers for radon have been designed based on scintillation counting but none are in general use.

Sensitivity of these instruments is mainly a function of the sensitive volume and the instrument background. As none of the instruments are specific for a given gas, they are subject to interference from other radionuclides which can compromise sensitivity and accuracy. Moreover, except for alpha scintillation detectors, they are sensitive to external radiation. This source of interference is usually shielded against or compensated by a second, background detector, as appropriate.

Thus, reliability, accuracy and sensitivity of these instruments depend to some extent on local environmental conditions and optimum performance is achieved when only one radionuclide is present. A number of modifications of these basic instruments have been reported to reduce some of the interference and although not available commercially, the instruments can be constructed from information given in the literature.

4.2.2 Accumulative Air Samplers may be used for virtually any kind of radioactive air contaminant, including gases. With the exception of automatic samplers which will be discussed later, accumulative sampling yields concentration after a delay, usually on the order of hours to days. For the measurement of gross concentration, filter papers are used for particulate radionuclides, whereas absorbers and containers are used for gases. In all cases, the collector must be compatible with the analytic method.

4.2.2.1 Filter media. In the selection of filter media, the most important characteristics are filter area, resistance, and collection efficiency. A sufficient variety of filters is available with a range of these characteristics adequate for almost any practical situation.

Filter area in itself is an important criterion because for optimum counting performance, it should approximate the size of the detector. But it also affects sample flow rate because pressure drop across the filter is inversely proportional to area and directly proportional to resistance (see 4.4 Pumps). Thus, resistance, too, is a critical property in the selection of a filter. Collection efficiency, of course should be as high as possible, ideally 100%, although efficiencies of 95% or more are generally satisfactory for routine work. Efficiency is a complicated function of particle size, air velocity and of the detailed characteristics of the filter itself. In some filters it may be quite low at certain combinations of these factors but for routine air sampling there is an adequate variety of available filters having inherent efficiencies in excess of 99% to satisfy most requirements. Therefore, there need be no compromise in efficiency except in very rare circumstances. Table 1 is a list of representative air sampling filters and their performance characteristics.

Collection efficiencies in the table, expressed in terms of penetration, i.e., 1-efficiency, differ markedly at the different filtering velocities listed. Consequently, filtering velocity can be critical in deriving satisfactory performance from certain filters. Collection efficiency also varies markedly with particle size, exhibiting a minimum at a particular size that varies with filtering velocity. The size of the test aerosol, 0.3 μm , on which the listed efficiencies are based, is fairly close to the most penetrating sizes for normal ranges of sampling conditions. Finally, collection efficiency tends to improve with filter loading during sample collection. Therefore, collection efficiencies, in general, will be greater than indicated in the table, particularly at particle sizes both larger and smaller than 0.3 μm .

TABLE 1

CHARACTERISTICS OF REPRESENTATIVE AIR SAMPLING FILTERS^(7,8)

Type	Designation	Percent Penetration of 0.3 μ m DOP Aerosol				Pressure Drop	Ash	Source
		14.2 cm/sec	26.7 cm/sec	53 cm/sec	106 cm/sec	cm H ₂ O at . 50 cm/sec	Content (%)	
Cellulose Fiber	Whatman 1	14	7	0.95	0.061	110	.13	H. Reeve Angel and Co.
	41	34	28	16	2	46	<.10	
	541	66	56	40	22	39	<.10	
	IPC 1478	80	90	90	90	1.9	.12	Institute of Paper Chemistry
Glass Fiber	MSA 1106B	0.042	0.068	0.048	0.022	39	99.6	Mine Safety Appliances Co.
	Gelman A (no organic binder)	0.015	0.019	0.018	0.011	42	98.4	Gelman Instrument Co.
	Gelman E (organic binder)	0.030	0.036	0.030	0.014	36	98.1	
	Whatman GF/A	0.014	0.018	0.015	0.006	37	99.0	H. Reeve Angel and Co.
	Reeve Angel 934AH	0.009	0.010	0.006	0.003	48	99.5	
Mixed Fiber	H&V H-70, 9 mil cellulose asbestos	2.9	1.8	0.8	0.20	82	21.0	Hollingsworth and Vose Co.
	Type 5G, cellulose glass	30	32	32	26	6.4	8.1	
Plastic Fiber	Microsorban, polystyrene	0.20	0.24	0.23	0.14	27	<0.10	Delbag-Luftfilter
Membrane	Millipore*† AA, 0.8 μ m pore diameter	0.010	0.015	0.020	-	180	<0.10	Millipore Corp.
	Metrical*† GA-4, 0.8 μ m pore diameter	-	-	-	-	-	-	Gelman Instrument Co.
Nuclepore	Nuclepore,* 0.8 μ m pore diameter	-	-	-	-	127	0.7	General Electric Co.

*Available in wide range of pore diameters.

†Available in variety of compositions.

Detailed characteristics of available filters and their properties may be found in NRL Report 6054 by Lockhart and Patterson⁽⁷⁾ and in Air Sampling Instruments, published by the American Conference of Governmental Industrial Hygienists.⁽⁸⁾

Other properties of filters that may warrant consideration are hygroscopicity, absorption (of radioactive emissions), ash content, interfering elements, background radioactivity, fragility and cost. Hygroscopicity can lead to excessive pressure drop in humid atmospheres but this only becomes serious under conditions of near-saturation, as in a mine. (It can be an important factor in gravimetric analysis but this is rarely performed in the monitoring of radionuclides.) Absorption, the attenuation of decay emissions from radionuclides which penetrate deep into a filter, can be a substantial source of counting error in the measurement of alpha or weak beta emitters. This difficulty is negligible in highly efficient filters having relatively smooth, hard surfaces. Ash content and/or the presence of interfering elements are of concern in the case of chemical analysis. In direct counting, trace radioactivity in the filter medium constitutes an interference that can be critical at low counting rates. Many media exhibit no detectable background. Fragility and cost are practical, if relatively minor, considerations.

4.2.2.2 Filter holders. Criteria for filter holders are simple but critical. For the collection of work room air samples, filter holders should be open-faced such that sample air is drawn directly onto the filter surface from the atmosphere without passing through a tube, orifice or other obstruction. This precludes loss of the radionuclide to surfaces upstream from the filter. Obviously, this requirement can not be followed with in-line filter holders that are used for stack sampling although significant sample losses do occur in those systems.

According to present theory⁽⁹⁾ the diameter of the filter opening should be held within prescribed limits, depending on flow rate, to avoid biasing particle collection by the effects of sedimentation and inertia. For the range of more commonly used flow rates, 5 to 50 l/min, the theory indicates proper diameters to be on the order of 2 to 5 cm which are consistent with sizes that are likely to be selected to satisfy other criteria. However, the theoretical treatment applies to calm air, which is atypical of most occupational environs, and it has not been confirmed experimentally.⁽¹⁰⁾ Therefore, these biasing effects have not been shown to be of practical significance in routine circumstances.

The filter should receive adequate support so that it is not stretched or torn by the pressure drop caused by the flow of sample air. The filter holder should be free of air leakage around the filter as well as into or through the holder's component parts. Finally, filter changing and holder replacement should be convenient and positive.

Filter holders may be made of various metals or plastics. Generally, metallic holders are more reliable and durable.

4.2.2.3 Other collection methods. Gases may be collected by dry adsorption, typically with activated charcoal or silica gel, or by accumulation in a rigid container (flask, tank) or a plastic bag.

Activated carbon at room temperature is used for gaseous iodine and at reduced temperature for the noble gases argon, krypton, xenon and radon. In the usual procedure, sample air, after a filtration stage to remove particulate contaminants, is drawn through a tube or cartridge containing fine-mesh activated charcoal. Collection is highly efficient until break-through occurs, a predictable phenomenon that depends on sample flow rate, sampling period, and the gas's adsorption coefficient, and the concentration of other adsorbable gases. Following collection, the tube is sealed and counted.

Good sensitivity can be achieved for iodine with a small cartridge, about 1 cm in diameter and 3-4 cm long containing a few grams of 12-30 mesh activated charcoal. The cartridge may be made of plastic.

For noble gases the charcoal is chilled during sampling to promote good collection efficiency. Metallic cartridges are used because of the low temperature.

Silica gel is used for sampling tritiated water in a similar manner. Air is drawn through a cylinder (typically 30 cm long x 3 cm diameter) containing a non-indicating silica gel. After sampling, the collected moisture is driven from the silica gel by heating and analyzed in a liquid scintillation counter.

As an alternative to adsorption techniques, noble gases may be collected in glass or metal containers for subsequent analysis. The sensitivity of this procedure is limited by the finite volume of gas that can be collected and, therefore, tends to be less than adsorbing from a gas stream but is adequate in given circumstances. If larger volumes are needed than are possible with rigid containers,

plastic bags can be used for collecting sample air, the bags being filled by means of air pumps. By employing a low pumping rate, the sample may be collected over a relatively long time period, to give a time-average concentration.

4.2.2.4 Automatic instruments that sample and measure continuously are variations of accumulative samplers that produce very nearly direct measurement of airborne radioactivity and are often used as alarms. Actually, the measurement is of material that has accumulated for a finite period, adequate sensitivity being attained by concentrating radioactivity on the filter as in manual sampling. Thus, there is an inherent delay of some minutes or more between the occurrence of a given atmospheric concentration and instrument response.

There are two general types of automatic samplers, one employing a static filter paper and the other employing a moving filter in the form of a tape. In some instruments, collection is by impaction on a solid surface rather than by filtration. In either mode of operation, the principle interference is from natural radioactivity, the solid decay products of radon and thoron gases. In the static filter sampler, air is drawn continuously through a fixed filter paper which is monitored continuously with an alpha, beta, or gamma detector. Detection of the radionuclide of interest depends on an incremental increase in counting rate significantly greater than the counting rate from natural radionuclides alone. In the moving filter type sampler, the filter paper is held over a sampling orifice and then, after a delay, positioned in front of a radiation detector. In the delay period, the activity from natural radionuclides diminishes somewhat by radioactive decay but here again, detection of the radionuclide of interest depends on a counting rate significantly greater than the background value. Accordingly, these instruments are relatively insensitive in most circumstances but sensitivity is usually adequate to indicate concentrations much in excess of MPCs.

4.2.3 Collectors for Measurements of Particle Size. In current practice, particle size measurements are made either by respirable fraction samplers which separate the respirable and non-respirable fractions of the aerosol or by instruments with which complete particle size distributions may be determined. The former measurement is by far the most common at present because of its greater simplicity.

Respirable fraction samplers, alternately known as two-stage samplers, utilize inertia or gravity to separate from the aerosol the fraction of particles small enough to reach and deposit in the lung. The first stage may be either a cyclone or an elutriator

although, in the U. S. nuclear industry, cyclones are used almost exclusively. The standard cyclone separates particles by size in accordance with a curve adopted by the USAEC.⁽¹¹⁾ This collection characteristic is such that 100% of unit density, spherical particles $\geq 10 \mu\text{m}$ diameter are collected and 100% of particles $\leq 2 \mu\text{m}$ penetrate. The second stage is a high-efficiency filter paper which collects all particles that pass through the cyclone. Size-selective samplers are available that operate from as little as 2 l/min (and can be used as personal samplers) to as much as 150 l/min.

Numerous experimental calibrations of these instruments have been performed, not all yielding consistent results. Moreover slight deviations in dimensions of the cyclone that may occur in manufacturing can distort intended collection characteristics. Therefore, the possibility of significant error should be recognized in applying these instruments to occupational monitoring.

The only practical field instrument currently available for determining particle size distribution is the cascade impactor although other techniques have been shown to be valid on an experimental basis. The cascade impactor consists of three or more collection stages in series through which sample air is drawn in jets at successively higher velocities and impacted on solid surfaces such that particles are separated in successively smaller sizes. The final stage consists of a high-efficiency filter paper which collects all particles that pass the last impaction stage. Particle size distribution is determined graphically or by calculation from the fractional distribution of radioactivity on the respective stages.⁽¹²⁾ Cascade impactors are available in a variety of sizes for sampling at rates from a few liters per minute up to 600 l/min. Also, models are available with as many as seven impaction stages.

Since the cascade impactor was introduced,⁽¹³⁾ numerous design variations have been proposed for improving performance or for application to special circumstances. Several basic types are available, the main variation being in the jet design: a stage may have a single or multiple circular holes or single or multiple slits. Circular jets produce sharper particle size selection whereas slit jets accommodate larger deposits of aerosol. However, the incorporation of multiple jets in each stage compensates for the more limited deposition capacity of circular jets.

Calibration of these instruments should be performed with the particular type of aerosol that is to be sampled if accurate size data are needed. Accuracy may be compromised by varying amounts of wall losses and other effects peculiar to this type of instrument. However, with reasonable care, approximate particle size data can be obtained.

4.3 Pumps

Air pumps are available in a sufficient range of sizes and types to meet virtually any air sampling requirement. The most important quality to be considered in pump selection is nominal flow rate, closely followed by the pump's flow-resistance characteristic, i.e., the dependence of flow rate on static pressure at the pump inlet. Size and power consumption are qualities that can be critical in the selection of portable instrumentation. Noisiness, durability, and maintenance requirements are also points to be considered.

Both positive displacement and centrifugal pumps are used for air sampling. In general, positive displacement pumps have low capacities, can operate at high static pressures, and exhibit moderate reductions in flow rate with increasing resistance. Centrifugal pumps produce high flow rates, can develop only low to moderate static pressures, and exhibit sharp reductions in flow rate with increasing resistance. The relatively flat performance curves of positive displacement pumps are desirable in air sampling applications, especially in filtration sampling in order to minimize the tendency of flow rate to diminish as filter resistance increases. However, at flow rates much in excess of 50 to 100 l/min, positive displacement pumps are cumbersome and except where uniformity of flow rate is an overriding factor, centrifugal pumps are preferred because of light weight and smaller size. But because of the low static pressures developed by these pumps, collectors having low resistance must be used. This requirement is met in filter sampling by using filters of large area.

Flow rate produced by an air sampler depends on both the performance curve of the pump and the resistance to air flow presented by sampling components, typically the collector, flow meter and valve. Selection of an air pump, therefore, involves matching the characteristics of the pump and the sampling train which are represented by the solid lines P and S in Figure 1. (The dashed lines labeled "positive displacement" and "centrifugal" indicate typical performance curves of the positive displacement and centrifugal pumps, respectively.) Flow rate of the system

Figure 1. Representation of pump and filter characteristics.

occurs at the intersection of the two lines. If resistance of the collector increases during sampling, as in the case of a filter paper, after a period of time it might assume the characteristic shown by line S^1 and the flow rate would be diminished to a value corresponding to the intersection of P and S^1 . By noting the respective intersections of S and S^1 with each of the two dashed lines, in turn, one can see that the relative reduction in flow rate for positive displacement pumps is much less than for centrifugal pumps.

Pump performance curves are usually available from manufacturers. Resistance of filter media can also be obtained from manufacturers but may be found in the literature.^(7,14) Data for some of the more common filters are given in Table 1. These are usually expressed as a pressure drop corresponding to a given linear velocity through the filter. The resistance curve can be approximated by assuming direct proportionality between resistance and flow rate. Resistance of other kinds of collectors, and of flowmeters and valves usually must be determined experimentally.

4.3.1 Low Volume Pumps. The sliding vane pump is the most popular among positive displacement types for low volume sampling which comprises flow rates up to about 100 l/min. This kind of pump is available in capacities from 5 l/min to well over 100 l/min, it has a fairly good performance characteristic, and is reasonably quiet and quite durable. Some models do not require lubrication, a useful feature when samplers are in frequent service.

Piston pumps are used to some extent but are far less popular because of comparatively greater size, noisiness, vibration and cost.

4.3.2 High Volume Pumps. Centrifugal pumps are generally used at sampling rates much in excess of 100 l/min where size, weight, or cost are important factors. The popular "high volume sampler", a class of centrifugal pump employing a two-stage air turbine, fills most of the sampling requirements in this range. These pumps can operate at flow rates from 300 to 2000 l/min, depending on inlet resistance and weigh on the order of 10 to 20 pounds. However, they tend to be noisy, an unwelcome characteristic in many applications.

In permanent monitoring installations having a number of collectors manifolded to a single pump, either the sliding vane pump or the lobe pump, another form of positive displacement pump, provides suitable service. In larger sizes these units can deliver the requisite high volume rates at high static pressures.

4.3.3 Personal Sampler Pumps. Diaphragm and piston pumps of special design are used for personal samplers, the main requirement being extremely small size and battery operation. The highest flow rates attainable with present models are 2 to 4 l/min. A typical weight including rechargeable battery is about one pound. The minimum operating period on a fully charged battery ought to be eight hours. The pumps attach conveniently to the belt.

A peculiarity of most of these pumps is rapid pulsation of the air flow which can affect the performance of some kinds of collectors (e.g., size-selective samplers) and distort the calibration of flowmeters. This characteristic can be neutralized with a pulse damper. Many of the newer pumps are equipped with this remedial device.

4.4 Flowmeters

Sample volume can be measured directly with an integrating volume meter but because meters of this type (wet-test, spirometer, gas, etc.) are bulky and expensive, they are rarely used. Most air sampling instruments are equipped with flowmeters which indicate the rate of flow; sample volume is calculated from average flow rate and sampling period. Flowmeters offer the advantages of economy, compactness, and durability. Also, it is usually desirable to determine instantaneous flow rate directly for adjusting sampling rate. Flowmeters, as a class, are inherently less accurate than integrating volume meters but their accuracy is adequate for all but the most exacting sampling requirements.

4.4.1 The Rotameter. The rotameter, or variable orifice meter, is the most versatile and widely used flowmeter for air sampling at rates from a few cubic centimeters per minute up to about 100 l/min. At higher flow rates, rotameters are considerably larger and more expensive than alternate instrumentation. In the rotameter, flow rate is indicated directly by the height of a float in a vertical, calibrated, tapered tube. The instrument can withstand considerable mechanical abuse and is relatively trouble-free if care is taken to prevent the entry of dirt. The condensation of moisture within the tube is another operational difficulty that causes error.

4.4.2 Fixed Orifice Meters. Fixed orifice meters, also known as flat plate orifices, are simple and inexpensive to fabricate and at higher flow rates (≥ 100 l/min), they are far more compact than rotameters. Therefore, they are often used with home-made sampling equipment and they are generally favored over the

rotameter in high volume sampling. In fact, fixed orifice meters are integral parts of a number of types of commercial high volume sampling instruments. In the traditional fixed orifice meter, pressure drop across the orifice is measured by a liquid manometer but in air samplers, mechanical pressure gauges of various types are substituted for simplification and convenience. The dynamic range of the fixed orifice meter is less than that of a rotameter, a disadvantage in some applications.

4.4.3 The Critical Orifice. The critical orifice is a useful device, applicable to any flow rate, where a uniform air flow is essential. The important characteristic of this device is that flow rate is constant and independent of pressure drop provided that the downstream pressure is ≤ 0.53 times the upstream pressure and the upstream pressure is invariant. This is an inexpensive and reliable means of regulating flow rate in any application where the resistance of the collector (or detector) is constant. It is not particularly suited to filter sampling where dust loading can increase filter resistance significantly during the sampling period.

4.4.4 Flowmeter Calibration. Inaccurate sample volume is one of the most common and serious kinds of error in air sampling. Therefore, reliable measurement of flow rate is crucial.

It is advisable to check calibrations provided by manufacturers. This may be done by operating the flowmeter in series with a standard volumetric metering device such as a spirometer or a wet-test meter. It is important to be aware that manufacturer's calibrations are for standard temperature and pressure, unless noted otherwise. This leads to another consideration in the use of flowmeters. Since the flowmeter is usually positioned downstream from the collector in a sampling train, air density at the flowmeter is less than ambient and, therefore, at non-standard conditions. The deviation from standard conditions is even greater if the sampling equipment is operated at an elevation substantially higher than sea level. Flowmeter calibration should be performed at the air density of intended operation. Most flowmeters conform to the laws of aerodynamics and it is possible to recalibrate at different air densities by calculation. Figure 2 indicates applicable equations. Nevertheless, it is preferable to recalibrate empirically, as well, at least in the case of rotameters, some of which tend to deviate from theoretical predictions.

1. Constant rotameter reading: $A_s = A_x$

2. Constant volumetric flow rate: $Q_s = Q_x$

3. Constant mass flow rate: $Q_s \rho_s = Q_x \rho_x$

A = rotameter scale reading

ρ = air density

Q = volumetric flow rate

s = standard conditions

M = mass flow rate

x = other conditions

h = pressure differential

(For illustration in diagrams 2 and 3, ρ_x is assumed $< \rho_s$)

*For orifice meters.

Figure 2. Density corrections.

4.5 Precautions

Sampling instrumentation, whether home-made or manufactured, is subject to malfunction, wear, and improper use that results in measurement errors. Many of these difficulties are unique to particular instruments and can only be detected and corrected through experience but a few recurrent difficulties can be mentioned for emphasis.

An extremely common error is in sample volume. This may arise from flowmeter miscalibration or from air leaks. Sources of error in flowmeter use are covered in 4.4.4. . Any leakage upstream from the flowmeter, assuming the normal arrangement of components, causes error. The filter or detector holder and interconnecting tubing and fittings should be checked when sampling instrumentation is first acquired and periodically thereafter, perhaps annually.

Filter holders unsuitable for particulate sampling are often used, resulting in sample losses ahead of the filter or other biases. This is covered in 4.2.2.2.

Finally, inappropriate filters often are selected, probably because of the large variety of filters that are available. The important considerations are adequate collection efficiency for the particular air contaminant of concern, resistance that is not excessive with respect to the pump and desired sampling rate, and compatibility with the analytical method.

5. SAMPLE ANALYSIS

5.1 Types of Measurement

Analysis of radionuclides in air samples is nearly always performed radiometrically because of the excellent sensitivity and selectivity afforded by this mode of detection and because the results are obtained directly in the units of applicable concentration guides. The only important exceptions are natural uranium and thorium which are analyzed fluorometrically in some cases.

In frequent circumstances, there is but a single, known radionuclide in a given work environment and the requisite analysis is simply the quantity of radionuclide collected in air samples. Radiation counters are used for this purpose. In more complex circumstances, mixtures of radionuclides in varying proportions may be collected simultaneously and analysis must be both quantitative and qualitative. Individual radionuclides may be distinguished by their half-lives, types of emission, or emission energies. Chemical separation is often applied in combination with these measurements.

Most air samples are collected on filter papers because of the convenience in both collection and analysis. Filter papers are easily handled and stored, if necessary, the likelihood of contaminating counting instruments is slight, and the flat, uniform sample deposit is conducive to good, reproducible measurement efficiencies.

Gaseous radionuclides necessarily are collected in flasks or on absorbers from which the sample is transferred to analytical equipment in the laboratory. Exceptions are radon and thoron which may be measured approximately by sampling their solid decay products on filter paper.

5.2 Requirements for Counting Instruments

The principal criteria of instrument performance for optimal sample counting are good detection efficiencies for radioactive emissions of interest, discrimination against interfering emissions, low counting background, and stability with time.

Counting efficiency combines the purely geometrical consideration of the fraction of emissions from the sample intercepted by the detector and the fraction of intercepted emissions that is converted to an electrical signal. There are wide, inherent

differences in efficiency reflecting the basic interactions of radiation with matter and detector characteristics. Thus, because of the penetrating ability of gamma radiation, counting efficiency is generally low but a solid detector can be more efficient than a gas-filled detector because of its greater mass. Conversely, alpha detectors usually have fairly high efficiency (50% is common) because alpha particles are readily absorbed in very little mass. Very high efficiencies approaching 100% can be attained for alpha and low energy beta radiation by placing samples inside gas-filled detectors or liquid scintillation counters.

Discrimination against interfering radiation arising either from the sample itself or externally can be achieved by shielding, by the electronic circuit, or by selecting a detector of suitable properties. Shielding is used to reduce environmental radiation in beta and gamma counters. The counting circuit can be designed to select or reject signals on the basis of energy or coincidence. Some types of detector are selectively efficient by nature; a thin scintillator is efficient for alpha or low energy beta but insensitive to gamma radiation.

The background counting rate should be low compared to sample counting rates. Counting backgrounds, which, like counting efficiencies, are inherently variable among different detectors, depend upon the counter's discrimination as well as circuit noise and other factors. Generally, the backgrounds of alpha counters are low, on the order of counts per hour, whereas the backgrounds of beta and gamma counters are fairly high, on the order of counts per minute or per second. However, these inherent background rates are more or less compatible with counting requirements which are much more stringent for alpha-emitting nuclides because of their low MPCs than for beta and gamma emitters.

The efficiency and background of a counting instrument must remain stable with time to facilitate reproducible, accurate analytical results. This characteristic is mainly dependent on electronic circuitry but also can be influenced by noise or poor regulation in line power, instrument contamination, and improper instrument settings.

5.3 Types of Instrument

5.3.1 Ionization Chambers. In the simple ionization chamber, relatively low voltage is applied between two electrodes in an enclosed volume of air or other gas. Radiation in the gas volume produces gas ions and electrons which migrate to the electrodes, constituting an electric current that is measured with an electrom-

eter. Current is proportional to the intensity and energy of the radiation but is impractically small at low energies or intensities. Ionization chambers may be sealed or open to the air. Their operation is very stable with time.

By operating ionization chambers in the pulse mode, sensitivity with respect to both energy and time is greatly improved. Pulse chambers detect and count individual emissions whereas current chambers measure average radiation flux.

5.3.2 Proportional Counters. The magnitude of the pulses in an ionization chamber can be amplified tremendously by operating with higher voltage applied to the electrodes. In this proportional region, the pulse size remains proportional to the energy of the triggering radiation but sensitivity is greatly improved over the pulse ionization chamber. By the use of appropriate gas mixtures and external circuitry, pulses are terminated rapidly, permitting high counting rates without coincidence loss. Both sealed and gas-flow chambers are used.

5.3.3 Geiger Counters. The Geiger counter is a gas-filled ionization chamber that is operated in a voltage region higher than proportional counters where the pulses produced are very large and of uniform size, independent of the incident radiation energy. Because of the large pulses, electronic circuitry is simple and stable but the counter is limited in its counting rate and is incapable of energy discrimination.

5.3.4 Scintillation Counters. Radiation produces light flashes in solid and liquid phosphors which may be converted to electrical pulses and amplified by a photomultiplier tube. This is the principle of the scintillation counter which, by proper combination of type and size of phosphor and the counting circuit, may be used for measuring virtually any radionuclide. It is capable of very high counting rates but also it can be made very sensitive. It finds wide application in gamma spectrometry.

5.3.5 Semiconductor Detectors. In a manner somewhat analogous to the production of ionization in a gas, radiation generates a small current in certain crystals that may be collected and measured by means of an imposed electric field and a suitable circuit. Typical semiconducting crystals are silicon and germanium. Semiconductor detectors are capable of extremely high counting rates and very precise energy resolution. A primary use is for alpha and gamma spectrometry. Present crystals are limited to fairly small sizes.

5.4 Application of Instrumentation to Sample Analysis

5.4.1 Alpha Counting. Alpha disintegrations can be counted with ionization chambers, proportional counters, scintillation counters, or semiconductor detectors. Present use of ionization chambers is limited primarily to gases that are mixed directly with the counting gas inside the (pulse ion) chamber. Open window, gas glow proportional counters are reliable and effective for alpha counting but the frequent replacement of the gas supply is disadvantageous. Scintillation counters (using zinc sulfide) are probably in widest use, because of the favorable combination of low background, good efficiency, and good discrimination against beta and gamma radiation. Semiconductor detectors have less sensitivity because of higher background and lower efficiency.

5.4.2 Alpha Spectrometry. Alpha spectra are measured almost exclusively with semiconductors. For ultimate resolution, counting is performed under vacuum although some measurements can be performed satisfactorily at ambient pressure.

5.4.3 Beta Counting. Thin-window Geiger tubes, gas-flow proportional counters, and scintillation counters are used for beta counting. Geiger tubes are limited with respect to higher counting rates and (because of window density) to lower energies. For the very lowest energies, exemplified by C-14 and H-3, liquid scintillation counting is employed.

Beta spectrometry, as such, is not feasible but energies can be determined by successive counting with absorbers of different thickness introduced between the sample and detector.

5.4.4 Gamma Counting. Any of the aforementioned types of counters may be used to measure gamma radiation in air samples. However, scintillation and semiconductor detectors have faster resolving times and better counting efficiencies which are necessary in some circumstances. On the other hand, scintillation and, especially, semiconductor counters are more expensive than other classes of counters and, for that reason, are not usually chosen for this application unless their improved performance is required.

5.4.5 Gamma Spectrometry. Scintillation and semiconductor detectors are also used for gamma spectrometry, semiconductors surpassing the former in popularity because of their better resolution.

5.5 Precautions

The intricacies of sample analysis are beyond the scope of this manual but a few of the more prevalent considerations can be mentioned here.

The naturally-occurring radionuclides, radon and thoron and their decay products, present in all atmospheres, tend to interfere with radiometric analysis unless they are given time to decay after sample collection. Radon daughters, which are much more abundant than thoron daughters in most areas, decay with an effective half-life of about 30 minutes and a counting delay of three hours is adequate. Thoron daughters decay with an effective half life of 10.6 hours and where they exist in significant concentrations, a counting delay of several days is advisable. The presence of either kind of radionuclide on a filter can be detected by re-counting two or three times at intervals of a few hours.

The sensitivity of any counting method depends primarily on the background count rate of the counting instrument and estimates of low radionuclide concentrations can be seriously in error if the counting background isn't known accurately. Even in stable instruments for which the background count may be quite constant, a daily check is advisable because of the possibility of contamination from sample material. Background counts should be made with a blank filter in place because some filter media contain trace amounts of radioactivity.

Counting instruments also require standardization periodically. Standard sources used for this purpose should match the samples both in size and energy.

The contaminated or upstream sides of filters collected in clean atmospheres are often difficult to identify and some convention that is understood by the analytical staff should be followed by sampling personnel to assure that the proper sides of filters will be counted. This may consist of marking the exposed side of the filter or placing the filter in the sample envelope consistently with the exposed side toward the identifying number or label on the envelope.

6. PROTOCOL

6.1 Characteristics of Work Atmospheres

The dominant characteristic of occupational environments that influences air sampling protocol is variability. Concentrations of radionuclides vary enormously with location and time. Also, the physical and chemical properties of airborne radionuclides such as particle size and solubility that influence hazard tend to vary with processes. Consequently, sampling must be repetitive and extensive to characterize the environment adequately in terms of averages, ranges, peaks, and other parameters.

The main cause of variability is the multiplicity and intermittency of sources contributing to air contamination. Typical laboratories have a number of hoods or glove boxes from which radionuclides may escape, depending on the work being performed at any particular time. Processing plants contain numerous operations or items of equipment from which contamination escapes, again the frequency or duration depending on the work cycle. Viewed from the perspective of a large work area, each location of release is virtually a point source with a steep concentration gradient surrounding it. Thus, a large range of concentrations exists in the vicinity of even a single source. Superposition of multiple sources, ventilation drafts, and moving personnel creates a complex, continuously shifting distribution of concentrations.

Varying concentrations very often correlate with manual operations. Manipulation in a hood or operation of processing equipment is likely to release contamination, especially in the immediate vicinity of the worker. Therefore, the exposure of an individual worker may differ markedly from general conditions in the work room, being comprised mainly of a series of localized, high concentrations that the worker generates in the course of his duties.

The ranges in concentration that may exist with time and location depend on specific circumstances but factors of 10 to 100 are common and greater ranges are possible. The greatest differences occur in the vicinity of sources. The concentration directly at a hood or other source where a man is working is typically 10 to 100 times greater than the concentration a few meters away. Fluctuations tend to be less in general room areas away from sources although room concentrations may be 2 to 10 times greater during periods of peak work activity than in dormant periods.

In addition to these hourly and daily fluctuations, long-term variations occur in most installations. Seasonal trends are often noted, higher concentrations occurring in the cold season when doors and windows remain closed. Changes in production rate, experimental procedures, or equipment also influence levels of air contamination.

In general, the degree of variation in air concentration is greater for particulate than for gaseous contaminants. The mechanisms for the release of gases to the work environment are less dependent on manual operations, gases diffuse more rapidly in air, and gases are unaffected by many of the forces (gravitational, inertial, thermal, and electrostatic) that influence the distribution and deposition of particles. Nonetheless, variation in gas concentrations can be great and must be taken into consideration in air sampling.

Recognition of these complexities is important in establishing an air sampling program. Basic considerations are types of sample, location and frequency of sampling, duration of samples, and interpretation.

6.2 Types of Sample

6.2.1 General Air (GA) Samples. GA samples are intended to reflect concentrations over broad areas and the exposures of personnel when not directly engaged in contaminant-producing activities. They tend to be the most useful, reliable, and simple of the four kinds of air sample and, consequently, constitute the backbone of most monitoring programs. Sampling instruments may be either permanently installed or portable but they remain in fixed locations during sampling. Sampling locations are in normally-occupied work areas, usually away from sources of localized air contamination and always away from walls or large obstacles that might cause anomalous contamination levels. The ideal height of collection for exposure evaluation is 5 to 6 feet from the floor. In corridors and busy work areas, samplers are placed overhead (preferably not higher than 8 feet) or at the sides of the areas. Samples are collected for fairly long periods, from 30 minutes to eight hours or more, so as to obtain time-average concentrations except in the case of radionuclides with half-lives that are short enough to influence sampling period. For example, an 8-hour sample of radionuclide with a 30-minute half life will indicate, primarily, the concentration in the last hour or so of sampling and will have no greater sensitivity than a 3-hour sample.

Flow rates used for GA sampling are from a few liters/min to about 1 m³/min.

6.2.2 Breathing Zone (BZ) Samples. Breathing zone samples are collected to measure the localized, transient concentrations to which workers are exposed while engaged in specific, contaminant-producing operations. In order to represent exposure, the sample must be collected close to the subject's nose because of the sharp concentration gradients around most sources; large differences in concentration may occur over distances of only a fraction of a meter. But a compromise in the ideal sampling position is necessary to avoid interfering with the worker's activities. For manual BZ sampling, used when the subject moves extensively during the operation, the sampler is held at the side of but always near the subject's face. For fixed-position BZ sampling, applicable to work at glove boxes or large machine tools, the sampler is situated a few inches above and forward of the subject's head.

The sampling interval matches the time of the operation. Untended, fixed-position BZ samplers may be activated automatically by a photocell or other sensors whenever a worker occupies the operating position; these must be equipped with a sample volume meter or a cumulative timer for the calculation of concentrations.

The upper limit of flow rate for manual BZ sampling is about 30 l/min although portable high volume samplers with flow rates up to 600 l/min have been used. Flow rates for fixed-position sampling are similar to those used for GA sampling.

6.2.3 Personal Air (PA) Samplers. Conceptually, PA samples are equivalent to continuous BZ samples. They are collected with compact, self-contained samplers worn by the subjects. The sample holder is attached to the front of the subject's torso. The sampler may be operated for a full shift to obtain a daily average exposure or it may be used to measure BZ concentrations at specific locations or operations of interest.

Sample flow rates of 2 to 4 l/min are typical for this kind of sampling instrument.

6.2.4 Process Samples. Process samples are collected specifically to identify or measure sources of air contamination and differ from GA samples only in that they may not necessarily reflect exposures to personnel. The process sample may be

collected in a normally inaccessible or prohibited location that is not a part of anyone's normal exposure. Process samples are usually collected in the proximity of equipment suspected to be a source of air contamination. Equipment and procedures are otherwise identical to GA sampling.

6.2.5 Precautions. As indicated in the preceding sections, sampler placement is critical but there are other special precautions that must be observed. Locations should be avoided where the sampler can be affected by direct contamination from the process or where the air concentration is abnormally high because of a localized effect, unless, of course, the local effect is being investigated. If a sampler is operated on a horizontal surface, as a convenient means of support, care should be taken that air discharged from the sampler is not directed at the surface, causing localized excessive air concentration from resuspended surface contamination.

Contamination of filters should be avoided by manipulating filter media with forceps and storing filters in envelopes or boxes before and after use. A common procedure for avoiding contamination is to preload a number of sample holders with filters in a clean area and carry the holders to the sampling area in a protective covering. Control filters should be collected periodically as a check on cleanliness. The control filter should receive the same treatment as sample filters including being mounted in the filter holder except that no air should be drawn through.

Sample flow rate may change with time in dusty atmospheres or during long sampling periods because of increased filter resistance. This effect should be compensated either by readjusting the flow rate periodically with a valve or by noting the start and end flow rates and taking the mean for calculating sample volume.

Vandalism may be a problem depending on local attitudes. Sampling instruments may be tampered with or turned off, filters may be punctured or contaminated or flooded with cigarette smoke, or other acts may be committed either malevolently or mischievously. The solution lies in ingenuity and remaining alert.

6.3 Monitoring General Air Concentration

At facilities where the exposures of employees ordinarily do not exceed 3/10 of the MPC, monitoring general air concentrations of radionuclides in work areas is considered to be adequate

(Section 2.4). This consists of simple measurements of total concentration without regard to particle size or other physical characteristics. Commitment to this kind of monitoring ought to be based on evidence that exposures are, in fact, low. In a new facility, a one-time survey of individual exposures (using methods described in the following section) or observing that substantially all air concentrations obtained in a comprehensive survey of work areas are less than 3/10 MPC constitute convincing evidence. Generally, it also would be a prerequisite that there be negligible opportunity for employees to be exposed to excessive local concentrations at manual operations.

After the inherently low order of air contamination is confirmed, the principal aim of monitoring is to assure that air concentrations remain at satisfactorily low levels. Toward that end, sampling must be performed periodically in all areas where there is a possibility of air contamination. The frequency of sampling depends on the stability of operations but because of seasonal effects, which are present in virtually any type of facility, quarterly monitoring is considered minimal.

6.3.1 Placement of Samplers. In the assumed circumstances, concentrations generally will be a small fraction of the MPC and monitoring should be arranged to disclose trends or frank excursions to higher concentrations. Ideally, samplers should be situated to reflect average concentrations in general work areas and to detect localized deterioration of control systems. For the former, samplers should be distributed in all normally occupied areas; for the latter, samplers should be placed in the vicinity of potential sources of release. These may be given preference on the reasonable grounds that they are the most sensitive indicators of incipient trouble but, typically, sources are distributed throughout a facility and sampling locations often may be chosen to serve both needs equally well.

The spacing among sampling locations can only be determined by experience at the particular facility. The primary consideration is that excessive concentrations will not be undetected. Areal variability of concentration is the key factor, the greater variation requiring closer spacing of sampling. The initial measurements by which it is determined that concentrations are low should constitute a reasonable basis for judging the spacing of subsequent samples. In general, spacing should be such that the standard error of the mean does not exceed 50% for samples covering a particular area of the facility.

6.3.2 Sampling Frequency. The frequency with which sampling is repeated depends on how rapidly conditions affecting air concentrations are likely to change. In a manufacturing facility with repetitive operations, these changes are apt to be infrequent enough for quarterly monitoring to be adequate. By contrast, an experimental facility is subject to frequent change, possibly necessitating continuous monitoring. Other important considerations are the likelihood and the consequence of undetected, high concentrations. A ten-fold increase in air concentration might occur in a plutonium facility with no overt signs whereas this would be extremely improbable in a natural uranium plant. Regardless of the selected frequency for routine monitoring, samples should be collected promptly whenever a new potential source of air contamination is introduced.

6.3.3 Types of Sampling Instruments. General area sampling may be performed with either permanently-installed or portable samplers, the choice depending on a number of factors. In a new facility it is preferable to avoid commitment to a given type of sampling equipment until the characteristics of air contamination are determined.

Permanent installations may consist of either a number of fixed, independent integral samplers or a series of collectors (usually filter holders) manifolded to a central, high capacity air pump. Permanent systems are initially more costly and often are difficult to install but require less man power in routine use, provide exactly reproducible sampling points, and tend to be favored for frequent sampling. Portable samplers are initially less expensive, require more man power in routine use, are more adaptable to changing conditions, and are preferable for infrequent sampling. Even where conditions warrant a permanent installation, it is advisable to have a few portable instruments available for special circumstances.

6.3.4 Data Interpretation. Air concentration data should be examined (a) to substantiate that essentially all exposures are $\leq 3/10$ MPC, (b) to detect trends that can't be accounted for by seasonal variation or other recognized conditions, and (c) to evaluate the sampling program for excesses or deficiencies.

Objective (a) can be achieved by a variety of methods. If relatively few samples are involved, a simple scan of the tabulated values will suffice. For sizeable numbers of samples and especially if some values exceed $3/10$ MPC, a listing by concentration intervals, a histogram of frequency versus concentration interval, or a cumulative plot of frequency distribution will organize the data and

facilitate interpretation. A plot of cumulative frequency distribution on probability paper shows directly the fraction of samples in excess of any particular concentration such as 3/10 MPC. It also yields directly the mean and standard deviation of the concentration values, two simple statistical parameters which characterize the entire group of samples.

Trends (objective b), which indicate changes in control effectiveness, can be followed by examining the averages of periodic groups of samples. Until some experience is gained, normal variations may be mistaken as signals of trouble and, conversely, abnormal trends may be obscured by natural variation. Organization of sample data by the methods mentioned for (a) or by a simple time chart of average concentrations will facilitate comparisons. In addition, simple statistical analyses are useful in determining if respective groups of samples differ significantly (Section 7). The primary use of this kind of analysis is to detect trouble in the form of rising concentrations resulting from deteriorating control. Both location and magnitude are important in this regard.

With respect to (c), the sampling program should be reviewed periodically to be sure that coverage is adequate but also to eliminate unnecessary sampling. For this purpose, a plot of cumulative frequency distribution is perhaps most informative because it indicates directly the probability that high concentrations are not being monitored. But other important considerations are physical changes in the facility, particularly enlargements, reductions, or re-allocations of space that may require adjustments in protocol.

6.4 Monitoring Employees' Exposures

Where exposures are likely to exceed 3/10 MPC, procedures should be implemented for monitoring individual employees. This is far more complex than monitoring general air concentrations because the workers' movements constitute an additional cause of variation that must be taken into account. The ideal (and only accurate) method of measuring an individual's average exposure is by the continuous collection of air samples directly in the breathing zone for an entire shift, work week, or other, designated averaging period. But this is impractical. Compromises with the ideal are necessary because of the limitations of man power and equipment. And it is generally conceded that safety is not jeopardized by a moderate compromise on accuracy, mainly because of the inherent imprecision and conservative nature of MPCs.

Conceptually, the personal sampler most nearly approaches the ideal means of monitoring individual exposures and for non-repetitive jobs, it is often the only method with acceptable accuracy. But personal samplers are not adaptable to every situation and do not, alone, provide all of the monitoring data required for engineering control. Their sample volume may not suffice for radionuclides having very low MPCs and they do not reveal the magnitude nor location of peak concentrations.

An alternate although less accurate method which is particularly suited to repetitive jobs consists of collecting both breathing zone and general air samples and logging the times spent by the worker at his respective assignments. With this information, his time-weighted exposure can be calculated and specific sources of exposure can be identified.

Satisfactory estimates of individual exposure can be based on general air samples representative of all work areas if contributions to individual exposures from local sources are insignificant and variations in general air concentrations are not severe.

Combinations of the foregoing methods are often applied to meet monitoring requirements.

6.4.1 Monitoring Exposures with Personal Samplers. The personal sampler is worn continuously by a worker with the sample holder positioned near or on the lapel. By sampling at a constant rate very near to the worker's breathing zone, it provides a fairly accurate measurement of average exposure if proper precautions are taken. Its preponderant mode of use is sampling radioactive aerosols by filtration but it may be used in conjunction with absorbers (e.g., charcoal) for sampling iodine or other gases.

Most personal samplers operate at nominal flow rates on the order of 2 l/min, yielding a sample volume of about 1 m³ in eight hours which is adequate for the measurement of most radionuclides. Improved sensitivity and a better average exposure can be obtained by sampling for an entire work-week but the longer sampling period increases filter resistance, tending to reduce flow rate, and increases the risk of compromising a sample by contamination or damage.

All employees in the $\geq 3/10$ MPC category should be monitored with personal samplers. For groups of employees performing identical work, it may be possible to characterize all exposures

by means of a single sampler worn by a representative of the group but this procedure should be adopted only after the separate exposures of all have been shown by measurement to be alike; individual work habits may cause substantially different exposures at identical tasks.

Sampling frequency depends on the magnitude and variation of exposures, information that is available only after the monitoring program is initiated. Generally, it should be adjusted so that quarterly averages can be determined to $\pm 50\%$.

The main precautions in the use of personal samplers concern contamination and the regulation of flow rate.

Location of the sample holder on the worker's torso renders the filter vulnerable to contamination from the worker's clothing and hands and from the process itself by direct contact or throw-off. Some filter holders are fashioned with protective screens or baffles, reducing the likelihood of contamination but also tending to interfere with aerosol collection. The need for a protective device can be determined by the use of controls. A dummy filter holder, identical to the actual holder but not connected to a pump, and fastened near the actual filter holder on the worker's torso, is equally subject to contamination. The amount of radionuclide, if any, detected on the dummy filter indicates the magnitude of contamination that might be present on the actual filter. If the amount is found to be small and reasonably consistent in repeated measurements, it may be treated as background and subtracted from the total amount of radionuclide measured on the actual filter. Excessive amounts of contamination indicate the need to relocate the filter holder, add some kind of protective device for the filter, or both.

Flow rate of the personal sampler varies with pressure drop across the filter which increases with dust loading. Over an eight-hour period, substantial loadings can accumulate in dusty atmospheres. Increased pressure drop can be compensated, within reasonable limits, by adjustment of the regulator valve, a standard feature of all personal samplers. Deviation from the nominal flow rate should not be allowed to exceed 10% during the sampling period. Readjustment several times during the shift may be required to achieve this criterion.

The quantity of radionuclide collected in the sampler divided by the total sample volume yields the average concentration to which the worker was exposed during the sampling period. Measurements should be repeated sufficiently in each period of three

months to obtain a precision of $\pm 50\%$ or better

Measurements by a personal sampler generally do not indicate sources of exposure. Exposures at specific operations or locations can be measured by operating the personal sampler only when an employee is at the location or job of interest but the inherent low flow rate of the instrument limits effectiveness in this application. Manually-collected breathing zone samples (section 6.2.2) are more suitable because of higher possible flow rates and they enable monitoring personnel to observe the suspected source of exposure and thereby supplement air concentration data with personal observation.

6.4.2 Monitoring Exposures with Breathing Zone (BZ) and General Air (GA) Samples. The average exposure of an employee can be calculated from measurements of all air concentrations normally encountered by the employee and a record of time periods spent at each location and task. Exposures derived by this procedure are less accurate than those obtained with personal samplers but sources of exposure are directly identifiable, an important advantage in maintaining control. The procedure ordinarily entails the manual collection of many samples per employee and therefore can be applied with only limited frequency. It is most adaptable to repetitive jobs.

If this procedure is used in place of personal samplers, it should be applied to all employees in the $\geq 3/10$ MPC category (section 2.4). For groups of employees performing identical work, it may be possible to characterize all exposures by measurement of only one person in the group but this procedure should be adopted only after the separate exposures of all have been shown by measurement to be alike; individual work habits may cause substantially different exposures at identical tasks.

Sampling frequency depends on the magnitude and variation of exposures and generally should be adjusted so that quarterly averages can be determined within $\pm 50\%$.

For each employee being monitored, BZ samples (section 6.2.2) should be collected at every manual operation which may be the cause of air contamination. Samples should be repeated several times for averaging because of the variability of air concentrations at manual tasks. Three is the usual number although many more may be necessary if the range of concentrations exceeds a factor of ten. The duration of the BZ sample should match the duration of the operation if possible. This assures that the sample will accurately

characterize the exposure and when properly noted in the sample log, gives time data needed for the exposure calculation.

General air samples (section 6.2.1) should be collected in all areas normally occupied by the employee, including change rooms and rest areas if air concentrations greater than background occur in such locations. The timing and duration of GA samples is primarily a matter of convenience although certain constraints apply in given circumstances. For accuracy in ascertaining exposures, samples should be collected only during periods of occupancy. Thus, in change rooms and lunch rooms where air concentrations are maximum during periods of occupancy, samples should be collected only in periods of use. Similarly, GA samples collected in work areas during lunch and rest periods may not be representative of exposures. Otherwise, the timing of GA samples in work areas is not critical unless routines follow a dominant cyclic pattern.

The minimal sampling period is dependent on analytical sensitivity (section 4.1.2). Generally, longer sampling periods yield better average values (with the exceptions noted above). Usually, the upper limit is the duration of a work shift. Duration may be influenced by the type of instrumentation used (section 6.3.3), longer sampling periods tending to be used in the case of permanently installed samplers.

The time periods spent by the employee at different locations and operations are obtained most reliably by direct observation but also can be ascertained by interviewing the employee or his supervisor or from time records.

Average (time-weighted) exposure is calculated by

$$X_{tw} = \frac{\sum x_i t_i}{t_i}$$

in which x_i is the average concentration at an operation or area and t_i is the corresponding time per averaging period (8-hour shift, work week, etc.) spent by the employee at that mode of exposure. An example of a job analysis sheet on which this calculation is made is shown in Figure 3.

The individual products ($x_i t_i$) indicate the relative contribution of individual areas and operations to the exposure which is useful in selecting remedial action in the event of excessive exposures. For example, temporary reduction in exposure could be

FIGURE 3

B-04-09

NY Form 144
(11-4-52)

JOB ANALYSIS SHEET

OPERATOR Saw Operator 1 MEN/SHIFT: 2 SHIFTS/DAY: 2 MEN/DAY

OPERATION OR OPERATING AREA	TIME PER OPERA. (MIN.)	OPERA. PER SHIFT	TIME PER SHIFT (MIN) (T)	NO. OF SAMP. LES	CONCENTRATION dis/min/m ³			AV'GE. CONC'N. TIMES TOTAL TIME (T×C)
					LOW	HIGH	(C) AVGE.	
BZ cutting tube samples on automatic saw	1.5	10	15	4	110	710	180	2.700
BZ cutting extruded tubes on automatic saw	8	10	80	3	620	1300	990	79,000
BZ stamping cut tube and rolling tube to storage conveyor	2	10	20	3	240	810	540	11,000
GA saw area			155	8	130	590	110	44,000
GA runout area			60	4	40	160	120	7,200
GA pickling area			60	6	12	34	19	1,100
GA tube storage area			60	6	33	150	76	1,500
GA locker room			10	2	75	160	120	1,200
GA lunch area			20	3	5	14	19	180

* Adjusted to two significant figures.

ΣT 480 $\Sigma (T \times C)$ 151,000

$\Sigma \frac{(T \times C)}{\Sigma (T)} = \frac{310}{480}$ Dis/min/m³ = 1.1 TIMES THE MAXIMUM ALLOWABLE CONCENTRATION

achieved by instituting respiratory protection at the individual tasks contributing most to the daily intake.

Some estimate of the accuracy of exposures calculated by this method can be deduced from the ranges of concentration making up the sub-averages (x_i). A tenfold range for a set of three samples has a mean standard error on the order of 50% which is generally satisfactory for minor components of exposure. For a $x_i t_i$ group that constitutes more than half of an individual's time-weighted exposure, the standard error of the concentration mean should be held to no more than 30%. This may require ten or more samples for a ten-fold range of concentrations and a greater number for broader ranges.

Errors also occur in the time intervals used in exposure calculations but ordinarily these are subordinate to concentration errors and contribute insignificantly to the composite error of the exposure value.

6.4.3 Monitoring Exposures with General Air Samples. At facilities designed for automated or remote operation, the localized, transient high air concentrations associated with manual operations are minimal and most exposures can be determined adequately by means of general air sampling. (Usually some breathing zone or personal air sampling is required for non-routine operations.) Calculations of exposure may consist of simple averaging of general air concentrations or may have to account for the stay-times of individual workers, depending on the variation of concentrations among work areas and the amount of worker movement.

Procedures are generally similar to monitoring general air concentrations (section 6.3). Samples should be collected in all occupied work areas so as to reflect exposures in all modes of operation, and in lunch rooms, change rooms, and rest areas if concentrations in excess of background may occur there. In the latter areas, only samples collected during periods of occupancy should be used for calculating exposures because of the reasons given in section 6.4.2. Preferred locations for samples in work areas are any work stations where employees tend to remain for long periods of time. As indicated in section 6.3, the spacing and frequency of sampling depends on areal and temporal variation of concentrations.

Calculations of exposures at the typical facility must take stay-times into account because of the differences in average concentrations among areas. To determine the necessity for this at a particular facility, average concentrations by area should

be compared. If averages do not differ by more than 50%, all workers' exposures can be assumed to equal the overall average concentration for the facility. But more typically, averages will differ markedly and exposures should be calculated for individual workers by the time-weighting method described in section 6.4.2 although in this case, breathing zone samples will not be involved. Interpretation of the exposure calculations is also similar to that described in section 6.4.2.

6.4.4 Particle Size Measurements. Instrumentation has been available for particle size measurements for many years (section 4.2.4). Undoubtedly, size measurements refine the information that may be used for assessing occupational exposures but size is only one of a host of variables that influence biological hazard. Moreover, nuclear facilities currently are operated in accordance with MPCs expressed in terms of total air concentration which, in the general case, are conservatively derived (section 2, 3, and 2.4). Therefore, there is little practical merit for incorporating these measurements in routine monitoring programs.

Nevertheless, particle size measurements are useful in special circumstances, two of which are accidental high exposures to very hazardous radionuclides and prolonged periods of exposure at concentrations near the MPC.

Unfortunately, particle size measurements are rarely obtained during accidents but if they are available, they can be useful in improving estimates of respiratory deposition and clearance rate. This information may contribute to the selection of proper treatment of serious cases of exposure or, more often, is used in calculations of dose commitment. In cases of suspected high body burdens it is often considered worthwhile to reconstruct the circumstances of the accident, with suitable controls, to obtain particle size and other measurements.

Where exposures are consistently (and unavoidably) near the MPC it is advisable to determine if sizes deviate markedly from the range for which MPCs are calculated, about 0.3 to 3.0 μm diameter. At sizes below that range, pulmonary deposition increases in relation to the assumed standard fraction and at larger sizes it diminishes. By contrast, tracheobronchial deposition, an additional factor in the radiotoxicity of soluble aerosols, increases at both smaller and larger sizes. Hence, doses from exposures at the nominal MPC might deviate significantly from those calculated for average conditions and, in fact, might be excessive.

The fraction of airborne radionuclide that deposits in the pulmonary region of the respiratory system can be measured directly with a respirable fraction sampler. This is the simplest field method available for the purpose. The measured fraction, after suitable adjustment, can be compared with the fraction assumed in MPC calculations (25%). The adjustment consists of multiplying the sampled respirable fraction by 0.326, in the case of cyclone samplers or multiplying by 0.312 and subtracting 0.012 in the case of elutriators.⁽¹²⁾ These corrections are for cyclones having characteristics defined by the "AEC" curve of respirable fraction⁽¹¹⁾ and for elutriators having a characteristic like the "BMRC" curve of respirable fraction.⁽¹⁵⁾ However, the respirable fraction sampler is directly applicable only to the assessment of long-lived, insoluble radionuclides for which the deep lung is the critical organ.

For soluble radionuclides and certain short-lived, insoluble radionuclides, deposition in the other regions of the respiratory system becomes an additional factor in dose assessment. Fractional deposition in the pulmonary, tracheobronchial, and nasopharyngeal regions can be determined by measuring the activity median aerodynamic diameter (AMAD) and referring to graphs prepared by the ICRP Task Group on Lung Dynamics.⁽²⁾ For the additional step of inferring dose resulting from either an accidental, one-time exposure or chronic exposure involves the use of available tables and graphs.⁽¹⁶⁾

The cascade impactor is the most practical field method for measuring AMAD. This instrument is more complex than the respirable fraction sampler, requiring four or more analyses per measurement and a graphical or mathematical treatment of the results. But this procedure yields both the AMAD and its geometric standard deviation.

Since the particle size distributions of aerosols are just as variable as aerosol concentrations, the placement of particle size sampling instruments is equally critical. Particularly in accidental releases, size characteristics are likely to vary greatly with time and with proximity to the point of release. Samplers should be operated as close to the exposed individuals as possible. In actual accidents, opportunities are rare for proper positioning of sampling equipment but some preplanning is desirable about the need for such measurements, availability of equipment (on standby, perhaps) and suitable procedures for implementation. Obviously, in reconstructions of accidents, measurements can be optimal with respect to time and location.

For routine exposures to insoluble radionuclides at concentrations near the MPC, a personal sampler equipped with a respirable fraction cyclone is the easiest and most accurate technique for obtaining particle size information. Where this instrument cannot be used because of its low flow rate of about 2 l/min, samples may be obtained by respirable fraction cyclones of higher flow rate, about 10 l/min, connected to hand-held air pumps and GA samples may be collected with similar instruments or samplers of higher flow rate up to about 150 l/min.

The cascade impactor is the preferred method of measurement for soluble radionuclides because of the aforementioned importance of size distribution in estimating radioactivity. Although available in a size small enough for use as a personal sampler, units of larger capacity are normally used in order to obtain sufficient sample volume. Models with sampling rates of 15 to 20 l/min are small enough to be used for breathing zone samples and the same units or larger ones can be used for GA measurements.

6.5 Emergency Monitoring

Procedures outlined in the foregoing sections are for normal operations in which radionuclide concentrations fluctuate around the MPC or below. In some circumstances there may be a need for special procedures applicable to accidental releases resulting in abnormally high radionuclide concentrations. If this occurs, the dominant requirement is to prevent harmful exposures to employees and, possibly, the public. Secondary considerations are acquiring data during the release for later calculations of exposures and monitoring during subsequent decontamination which, even after the emergency phase, is not a routine procedure.

The function of air monitoring in the prevention of harmful exposures is to warn employees to leave or take protective action. (The latter might include actuating alarms or devices related to releases within or from the building.) Instruments are available for this purpose (sections 4.2.1 and 4.2.3) but the specific situation should be examined to determine their need. In some cases, alarm monitors may be mandatory while in others, alternatives are preferable.

The need for any kind of warning device depends, first of all, on the real possibility of an acutely hazardous release that otherwise would not be detected. This, in turn, depends on the quantity and form of the radionuclide and the nature of the nuclear facility. Ultimately, this is a matter of judgement although it's always prudent to be pessimistic if the circumstances are doubtful.

Consideration of alternate means of detection is important because an air monitor may not be the most effective means of warning. There is a finite delay from the time of release until the contaminant reaches the monitor and the initial concentrations may be diluted substantially in this interval. Modes of detection that sense an incipient release or conditions conducive to a release are preferable. For example, the ventilation system, a point of control vulnerability in most nuclear facilities, can be equipped with an alarm actuated either by power failure or a loss of static pressure. Fire, which rarely can be dismissed as a threat to control, can be sensed promptly by heat or smoke detectors within hoods and other critical locations. The malfunction of process equipment, itself, may constitute an indication of possible release.

Visual observation of a release, where applicable, is the most reliable warning to the people who may be affected. Aerosol clouds at concentrations of 10 mg/m^3 or more are visible, especially in the normally clean work room air of nuclear facilities. Ten mg/m^3 is 60 times the MPC for natural uranium, 550 times the MPC for Th-232, and 830 times the MPC for I-129. Exposure for ten minutes at 10 mg/m^3 would be equivalent to 1.2 days, 2.3 weeks, and 3.4 weeks at the respective MPCs of these three nuclides. In the case of iodine, 10 mg/m^3 is ten times the threshold limit value based on chemical toxicity and would cause severe respiratory irritation to exposed individuals. Hence, the senses of sight or smell are adequate warning mechanisms for some radionuclides.

In contrast to the other cited modes of detection, air monitor alarms are specific for releases of radionuclides. And where there is a significant risk of otherwise undetectable releases of radionuclides having low MPCs and high specific activities like plutonium and Sr-90, the air monitor is necessary for adequate protection although even in this circumstance, sole reliance should not be placed on the air monitor because other indications of contaminant release, notably excessive surface contamination, have been found to signal trouble more promptly, on occasion.

The number and placement of alarm monitors should be selected to minimize the delay between the actual release of a hazardous concentration of a radionuclide and the activation of alarm signals. A practical strategem is to place alarm monitors near ventilation exhaust grills through which contaminants inevitably must flow. This is an effective arrangement for small rooms or for large rooms with proportionately large numbers of exhaust points, provided that

ventilation rates are not less than six changes per hour. An alternate arrangement is to distribute alarm monitors in the work room near points of likely release. The efficacy of specific locations can be tested approximately by noting the time and direction of travel of smoke released from a smoke tube at various points where contaminants may escape.

Alarm monitors may be set to give a signal when a prescribed concentration is exceeded or when a prescribed time-integrated concentration is exceeded. In either case, the setting usually is equivalent to a large multiple of normally-acceptable exposure because of limitations on sensitivity imposed by interference from natural radionuclides, radon and thoron daughters, and to avoid spurious alarms.

Setting the monitor to alarm at some multiple of the maximum concentration that would be expected in normal operations is the most straightforward procedure. The alarm in this case does not necessarily imply a hazard but it is a clear-cut indication of a serious malfunction in the control system that warrants prompt corrective action. Nonetheless, it would also indicate that precautions should be taken to prevent unnecessary exposures such as donning respirators and, in some cases, evacuation.

Setting the monitor to alarm at a time-integrated concentration is a more sophisticated procedure that recognizes that hazard is most directly related to the accumulation of inhaled radionuclides, i.e., the hazard from inhaling concentration C for ten minutes is equivalent to inhaling concentration $C/10$ for 100 minutes. To be effective, the setting of a time-integrated concentration must be substantially less than a truly hazardous amount, otherwise the alarm is too late. On the other hand, the setting must be substantially above any non-hazardous time-integrated concentration that may occur in normal operations.

Concentration data that may be needed after the fact for estimating exposures during an accidental release may be obtained at least in part, from the alarm monitor itself. Where alarm monitors do not adequately represent potential exposures, fixed air samplers that are activated by the alarm monitor can be prepositioned in representative work areas and maintained in standby status. Such samplers, of course, are useless unless they are checked periodically for proper operation and for filter cleanliness and integrity. Unless the risk of hazardous exposures is very high and local medical authorities deem exposure data to be useful for therapy or subsequent exposure management, supplementary air samplers need not be installed.

Particle size is an important property of the radionuclide in estimating respiratory deposition resulting from a hazardous exposure. As in the case of supplementary air samplers, collectors for the measurement of particle size that are activated by the alarm monitor can be situated in work areas but this need not be done unless advocated by medical authorities.

Monitoring of decontamination work following an emergency release is conducted to assure that appropriate protection is provided to the personnel involved and to obtain a record of exposures. Radionuclide concentrations can be expected to be both higher and more varied than in routine operations therefore requiring more intensive sampling and, to be useful as a day-to-day guide on protective measure, more rapid analysis.

12

7. STATISTICAL METHODS

Extreme variation of radionuclide concentrations with time and location has been stressed as a major complication in selecting a satisfactory monitoring protocol and in interpreting sample data. Judgement and experience are critical attributes in managing this problem but the behavior of the concentrations of air contaminants, fundamentally, is statistical in nature requiring treatment by statistical methods. At the present state of air sampling practice, statistical methods cannot substitute adequately for judgement and experience but they can be useful aids. A few basic statistical considerations are presented here.

The following discussions apply only to random errors. Systematic errors are assumed to be reduced to insignificance by calibration and check procedures described in the sections on instrumentation and analysis.

7.1 Measurement Precision

The confidence that can be placed in air sampling data begins with the precision of individual measurements of concentration. For example, it is useful to know that a fifty percent difference between two samples reflects an actual concentration difference rather than statistical uncertainty of the two measurements. In the usual measurement procedure consisting of two steps, sample collection and sample analysis, the measurement precision reflects the combined precisions of the separate steps:

$$V_m^2 = V_c^2 + V_a^2 \quad (1)$$

where V_m , V_c , and V_a are the coefficients of variation, respectively, for the total measurement, sample collection, and sample analysis.

The precision of sample collection is substantially constant for a given type of sampler, operating procedure, etc., whereas the precision of analysis often is dependent on the amount of sample collected and, for that reason, variable. Therefore, measurement precision itself is subject to some variation but the influence of V_a can be held to reasonable limits by collecting adequate sample volumes or by extending counting periods, as necessary.

Measurement precision can be determined experimentally by collecting and analyzing a series of replicate sets of samples, each set consisting of two or more simultaneous samples. After the samples are analyzed and the individual concentration is calculated from each sample, the measurement precision is calculated as the average coefficient of variation of concentrations within sets. If sets consist of pairs, the coefficient of variation may be estimated by

$$v_m = \frac{0.886}{n_R} \sum \frac{\Delta x}{\bar{x}} \quad (2)$$

in which n_R is the number of pairs (replicate sets), Δx is the individual pair difference, and $\bar{x}$ is the individual pair mean. Dividing by the mean, which yields coefficient of variation instead of the more customary standard deviation, normalizes the concentration variability that is apt to occur with time during the series of replicate samplings.

For three or more replicate samples in each set, the conventional formula for coefficient of variation may be used

$$v_m = \frac{1}{n_R} \sum \frac{1}{\bar{x}} \sqrt{\frac{\sum (x - \bar{x})^2}{n-1}} \quad (3)$$

in which x is concentration, $\bar{x}$ is the arithmetic mean of a set of replicate concentrations, and n is the number of samples comprising a set.

Measurement precision need not be determined routinely but it is useful as an occasional check on the quality of monitoring procedures. Measurement precision is manageable and ordinarily ought not to exceed 10 to 20%, an amount that is small when considered as a component of the precision in measuring widely fluctuating concentrations of a radionuclide. In the event of greater error, the cause should be ascertained by examining the two components of measurement precision.

Analytical precision is usually estimated on the basis of radiometric count and is calculated as the square root of the total sample count, C , assuming a Poisson distribution. Calculated in relative terms to be consistent with measurement precision, it is divided by C :

$$V_a = \frac{\sqrt{C}}{C} \quad (4)$$

When the sample count is not much in excess of counting background, the background count must be included in the calculation:

$$V_a = \frac{\sqrt{C + B}}{C} \quad (5)$$

(The background count must be for the same counting period as the sample count.)

There is no direct means of measuring the precision of sample collection but it can be estimated indirectly from the analytical precision and the overall precision of measurement:

$$V_c = \sqrt{V_m^2 - V_a^2} \quad (6)$$

Both components of the measurement precision should be minimized within reasonable limits but if one is about as low as can be attained, reducing the other component to a much lower value than the first will not improve the measurement precision, itself, substantially.

7.2 Sample Averaging

Averaging is one of the most simple and useful treatments of sample data. The arithmetic mean of normally-distributed, random samples collected in a work area represents the best estimate of the exposure of an individual in the area during the sampling period. (See 7.4 concerning log normal sample distributions.) Also, comparison of arithmetic means of samples collected at different times or locations is a useful indication of the relative degree of control or of trends.

The reliability of any average depends on the number of measurements on which the average is based and on their dispersion or coefficient of variation. The coefficient of variation of a group of samples is calculated by

$$V_x = \frac{1}{\bar{x}} \sqrt{\frac{\sum (x - \bar{x})^2}{n-1}} \quad (7)$$

in which x is the individual concentration, $\bar{x}$ is the arithmetic mean of the concentrations, and n is the number of measurements. V_m , the measurement precision, is a component of V_x but its contribution ordinarily is quite small.

The coefficient of variation of the concentrations is independent of the number of measurements from which it is calculated although V_x , the estimate of coefficient of variation, improves with increasing numbers of measurements. However, the coefficient of variation of the average concentration, called the relative standard error of the mean, does depend on the number of samples and is calculated by:

$$V_{\bar{x}} = \frac{V_x}{\sqrt{n}} \quad (8)$$

The relative standard error of the mean can be used as a criterion of the adequacy of sampling protocol, any error greater than .50 (50%) being considered unacceptable. Also, in its more customary form, the standard error of the mean, it may be used to determine if two average concentrations actually differ, when this is not apparent from the relative magnitudes, and if an average is significantly above or below a given MPC. This involves the use of Student's t -tests which are described in 7.3.

7.3 Comparing Average Concentrations

The Student's t -test is a simple method of determining whether the mean of a set of samples differs significantly from a reference concentration such as an MPC_a , or from the mean of another set of samples. It involves a comparison of the standard deviation of the mean of the group of samples in question with the statistical parameter, t , which is defined by the equation

$$t = \frac{\bar{x} - \mu}{S_{\bar{x}}} \quad (9)$$

in which $\bar{x}$ = arithmetic mean of the samples

μ = true mean, or value with which $\bar{x}$ is compared

$S_{\bar{x}}$ = standard deviation of the mean of the samples

The first step of the procedure consists of calculating the mean and standard deviation of the mean by

$$\bar{x} = \frac{\sum x}{n} \quad (10)$$

in which x is the value of an individual sample and n is the number of samples, and

$$S_x = \sqrt{\frac{\sum (x - \bar{x})^2}{n-1}} \quad (11)$$

and

$$S_{\bar{x}} = \frac{S_x}{\sqrt{n}} \quad (12)$$

The next step depends on the kind of comparison to be made. To consider a simple case, it is desired to know if the sample mean differs significantly from a fixed value such as a maximum permissible concentration. Using equation (9), the difference between the sample mean and the maximum permissible concentration is divided by the standard deviation of the sample mean to obtain t . This is the number of standard deviations by which the two concentrations differ and is a measure of the likelihood that the difference is statistically significant. To quantify that likelihood, one must enter a table of the t distribution such as Table 2, at the appropriate number of degrees of freedom ($n-1$) to find the value of t closest to the one that was calculated. The decimal fraction at the head of the column in which t is found expresses the likelihood that the sample mean and standard concentration actually differ. For example, if $n = 10$ and $t \sim 1.8$, there is a 95% chance that the sample and the standard concentration are not the same.

If the means of two sets of samples are to be compared, the procedure is similar but the equations are more complex. The means and standard deviations are calculated for both sets of samples by equations (10) and (11). Then a combined standard deviation of the mean is calculated by:

$$S_{\bar{x}_{1,2}} = \sqrt{\frac{S_{x_1}^2}{n_1} + \frac{S_{x_2}^2}{n_2}} \quad (13)$$

and t is calculated by:

TABLE 2
PERCENTILES OF THE t DISTRIBUTION

df	$t_{.60}$	$t_{.70}$	$t_{.80}$	$t_{.90}$	$t_{.95}$	$t_{.975}$	$t_{.99}$	$t_{.995}$
1	.325	.727	1.376	3.078	6.314	12.706	31.821	63.657
2	.289	.617	1.061	1.886	2.920	4.303	6.965	9.925
3	.277	.584	.978	1.638	2.353	3.182	4.541	5.841
4	.271	.569	.941	1.533	2.132	2.776	3.747	4.604
5	.267	.559	.920	1.476	2.015	2.571	3.365	4.032
6	.265	.553	.906	1.440	1.943	2.447	3.143	3.707
7	.263	.549	.896	1.415	1.895	2.365	2.998	3.499
8	.262	.546	.889	1.397	1.860	2.306	2.896	3.355
9	.261	.543	.883	1.383	1.833	2.262	2.821	3.250
10	.260	.542	.879	1.372	1.812	2.228	2.764	3.169
11	.260	.540	.876	1.363	1.796	2.201	2.718	3.106
12	.259	.539	.873	1.356	1.782	2.179	2.681	3.055
13	.259	.538	.870	1.350	1.771	2.160	2.650	3.012
14	.258	.537	.868	1.345	1.761	2.145	2.624	2.977
15	.258	.536	.866	1.341	1.753	2.131	2.602	2.947
16	.258	.535	.865	1.337	1.746	2.120	2.583	2.921
17	.257	.534	.863	1.333	1.740	2.110	2.567	2.898
18	.257	.534	.862	1.330	1.734	2.101	2.552	2.878
19	.257	.533	.861	1.328	1.729	2.093	2.539	2.861
20	.257	.533	.860	1.325	1.725	2.086	2.528	2.845
21	.257	.532	.859	1.323	1.721	2.080	2.518	2.831
22	.256	.532	.858	1.321	1.717	2.074	2.508	2.819
23	.256	.532	.858	1.319	1.714	2.069	2.500	2.807
24	.256	.531	.857	1.318	1.711	2.064	2.492	2.797
25	.256	.531	.856	1.316	1.708	2.060	2.485	2.787
26	.256	.531	.856	1.315	1.706	2.056	2.479	2.779
27	.256	.531	.855	1.314	1.703	2.052	2.473	2.771
28	.256	.530	.855	1.313	1.701	2.048	2.467	2.763
29	.256	.530	.854	1.311	1.699	2.045	2.462	2.756
30	.256	.530	.854	1.310	1.697	2.042	2.457	2.750

TABLE 2 (Cont'd)

df	t _{.60}	t _{.70}	t _{.80}	t _{.90}	t _{.95}	t _{.975}	t _{.99}	t _{.995}
40	.255	.529	.851	1.303	1.684	2.021	2.423	2.704
60	.254	.527	.848	1.296	1.671	2.000	2.390	2.660
120	.254	.526	.845	1.289	1.658	1.980	2.358	2.617
∞	.253	.524	.842	1.282	1.645	1.960	2.326	2.576

From "Operational Aspects of Radiation Surveillance",
 Division of Radiological Health, Public Health Service,
 U. S. Department of Health, Education and Welfare.

$$t = \frac{\bar{x}_1 - \bar{x}_2}{S_{\bar{x}_1, 2}} \quad (14)$$

Also, calculating the degrees of freedom is more complex:

$$df = \frac{\left(\frac{S_{x_1}^2}{n_1} + \frac{S_{x_2}^2}{n_2} \right)}{\frac{\left(\frac{S_{x_1}^2}{n_1} \right)^2}{n_1 + 1} + \frac{\left(\frac{S_{x_2}^2}{n_2} \right)^2}{n_2 + 1}} \quad (15)$$

The result of equation (15) can be rounded off to the nearest integer.

The use of the t table and the interpretation of the derived values are the same as in the preceding case.

It is important to note that the t-test is only a measure of probability; it does not provide absolute certainty. The level of probability that is acceptable for making decisions depends on the importance of the decision. Frequently, a ninety percent probability is satisfactory but 95% or even 99% probability may be sought in critical circumstances.

7.4 Log-Normal Distributions

The foregoing discussions and formulations concerning sample averaging apply to samples characterized by a normal or Gaussian distribution where values are distributed symmetrically around the mean value. Many air concentrations are normally-distributed but more often they are log-normally distributed. In the latter case, sample distribution around the central value is skewed in the direction of the higher concentrations. However, the logarithms of the samples are normally distributed, enabling the data to be handled by fairly simple mathematical relationships.

The most significant aspect of this distribution is that the best estimate of the mean concentration is obtained from the distribution parameters, the geometric mean, m_g , and the geometric standard deviation, σ_g , rather than from the arithmetic mean of the samples. The following equation applies:

$$\ln \bar{x} = \ln m_g + 0.50 \ln^2 \sigma_g \quad (16)$$

Thus, the arithmetic mean of a log-normal distribution always exceeds the geometric mean and should be used for estimating exposure.

For low geometric standard deviations the difference between arithmetic means calculated by equation (16) and by sample averaging are small and the extra effort required for the log-normal calculation may not be justified. For example, at a geometric standard deviation of 2 and a sample size of 6, the difference is about 8%. But at larger geometric standard deviations, differences are substantial.

Whether a particular group of samples is better characterized by a normal or log-normal distribution can be determined most easily by alternately plotting the sample values on normal and logarithmic probability papers. The concentrations are arranged in ascending order or tabulated into intervals. The cumulative percentages exceeding various concentrations are plotted. Whichever plot more nearly follows a straight line is taken as the type of distribution. Small numbers of samples are apt to yield ambiguous results, ten being about the minimum needed to identify a distribution and a larger number being preferable.

The plot of log-normal sample data on log-normal probability paper also enables the geometric mean and geometric standard deviation to be obtained by simple graphical means. The geometric mean is the concentration corresponding to the intersection of a straight line through the data and 50% on the scale of cumulative percentage. The geometric standard deviation is the ratio of the concentration at a cumulative percentage of 86.13 to the geometric mean concentration.

7.5 Determination of Number of Samples

Deciding on the number of samples to be collected in a particular work area or to estimate a particular exposure entails a compromise between the need for good precision of the results and the practical constraints of available instrumentation and manpower. Often, various rules-of-thumb are invoked and this practice is not without merit because such rules grow from the collective experience of numerous practitioners. Nevertheless, blanket rules, by nature, cannot be optimal in all situations and it is preferable to draw on statistical procedures in the decision-making process.

For normally-distributed concentrations, an estimate of the number of samples needed to achieve a given relative standard error of the sample mean can be made by re-arranging equation (8) which was given in section 7.2:

$$n = \left(\frac{V_{\bar{x}}}{V_x} \right)^2 \quad (17)$$

In this case, $V_{\bar{x}}$ is a pre-selected value that is considered to be the acceptable error in sampling results and can be set to any desired value. It should be noted, however, that the required number of samples increases rapidly as $V_{\bar{x}}$ diminishes with respect to V_x .

In the use of these equations, the coefficient of variation of concentrations must be known or estimated beforehand. Since coefficients of variation are apt to vary widely with locality, the procedure is essentially limited to situations where they can be calculated from prior sample data.

In the preceding equation and in equation (8), the relative standard error of the mean has a confidence limit of 68%. This implies that if the given sampling protocol were repeated many times in the same area, the errors of the sample means would be $\leq V_{\bar{x}}$, 68% of the time and $\geq V_{\bar{x}}$, 32% of the time. Errors of the mean can be calculated for higher confidence limits by inserting z , the normal deviate, yielding the more general form of equation (17):

$$n = \left(\frac{zV_{\bar{x}}}{V_x} \right)^2 \quad (18)$$

The values for z are 1.65 for a 90% confidence limit, 1.96 for 95%, and 2.58 for 99%. At the 90% limit, the error of the mean would be expected to be $\leq V_{\bar{x}}$, 90% of the time and $\geq V_{\bar{x}}$, only 10% of the time.

For log-normally distributed concentrations, the equation is modified slightly:⁽¹⁷⁾

$$n = \left[\frac{zV_x}{\ln(1+V_{\bar{x}})} \right]^2 \quad (19)$$

The usual concepts of mean and standard deviation (or coefficient of variation) relate to an assumption that the size of population from which measurements such as air samples are obtained is infinite. Thus, no amount of samples will ever yield a true mean although the estimate of the mean continuously improves as more samples are obtained. This assumption of an infinite population does not apply to all monitoring situations. For example, if the average concentration of a radionuclide were needed at a fixed location during a particular 8-hour period and if 30-minute samples were collected for this purpose, the sample number could not exceed 16. In this case, the population would be finite and if 16 consecutive 30-minute samples were collected, the error in the mean due to concentration variation would be nil; the only error would be that arising from measurement precision.

The circumstance of a finite population arises frequently because monitoring is often directed to specified time periods (a work-shift, work-week, quarter-year) and to particular locations or employees. The salient point is that fewer samples are needed to achieve a given relative standard error of the mean than in the case of an infinite population. This is reflected in the following modifications of equations (18) and (19). For normally-distributed samples:

$$n = \frac{N (zV_x)^2}{N V_{\bar{x}}^2 + (zV_x)^2} \quad (20)$$

and for log-normally distributed samples:

$$n = \frac{N (zV_x)^2}{N \ln^2(1+V_{\bar{x}}) + (zV_x)^2} \quad (21)$$

In both equations, N is the size of the finite population, which can be calculated by $N = T_t/T_s$, the ratio of the total time in a chosen monitoring period to the time period of an individual air sample.

Conversely, the standard error of the mean of samples collected from a finite population is less than from an infinite population as indicated by the following rearrangement of the preceding equations. For normally distributed samples:

$$V_{\bar{x}} = zV_x \left(\frac{N-n}{Nn} \right)^{\frac{1}{2}} \quad (22)$$

For log-normally distributed samples:

$$V_{\bar{x}} = e^{zV_x \left(\frac{N-n}{Nn} \right)^{\frac{1}{2}}} - 1 \quad (23)$$

7.6 Precision of a Time-Weighted Exposure

A calculation of time-weighted exposure, in accordance with section 6.4.2, draws on average concentrations derived from several different groups of samples, each having its own standard deviation, and incorporates measurements or estimates of stay-times, also subject to random errors. Therefore, the error of the time-weighted exposure is a composite of the errors associated with each of the components. The calculation of the composite precision is simple but too tedious for routine use (unless it is computerized). Nevertheless, an occasional check is desirable to insure that sampling protocol is adequate. The following equation applies:⁽¹⁸⁾

$$S_E = \frac{1}{T} [\Sigma (t_i S_{\bar{x}_i})^2 + \Sigma (\bar{x}_i S_{t_i})^2]^{\frac{1}{2}} \quad (24)$$

In this, t_i is the time period of an exposure characterized by the mean concentration $\bar{x}_i$, $S_{\bar{x}_i}$ and S_{t_i} are the standard deviations of the mean concentration and mean time, and T is the total time period over which the time-weighted exposure applies, i.e., 8 hours, 40 hours, etc. The units of t and T should be identical. Inasmuch as the values of t are often estimated or based on information supplied by others, their standard deviations must also be estimated.

7.7. Effect of Sampling Period on Sample Coefficient of Variation

Intuitively one would expect the observed variation of air concentration to diminish as sampling period is increased and this effect is expressed by the equation

$$\frac{V_{x_1}}{V_{x_2}} = \left(\frac{t_2}{t_1} \right)^{\frac{1}{2}} \quad (25)$$

which shows that the coefficients of variation determined for two different sampling periods are inversely proportional to the square roots of the sampling periods.⁽¹⁹⁾ This relationship suggests lengthening the sampling period as a convenient means of improving estimates of mean concentrations without increasing the number of samples. This can be verified mathematically by noting the dependence of $V_{\bar{x}}$ on V_x in equations (8), (22), and (23).

This is an effective strategy in some situations but not invariably so because equation (25) is predicted on the assumption that successive air samples are independent of each other. This assumption is often invalid in actual practice because air contamination may vary cyclicly during the work day and samples spaced closely in time with respect to the period of a cycle will not be randomly distributed. In that case, the reduction in the coefficient of variation is less than indicated by the equation and for a given commitment of total sampling time, a larger number of short-term samples may well be more beneficial in terms of reducing the mean standard error than a correspondingly smaller number of long-term samples. However, this generalization fails if the total sampling period that is to be committed is a large fraction of the averaging period because in the limit (wherein the entire averaging period is monitored) the imprecision of the average concentration owing to concentration variance is nil and the only source of imprecision is the measurement errors, as discussed in 7.5. Consequently, in this circumstance, a few, long-period samples would be the choice over more numerous short-period samples if convenience and efficient use of manpower is the dominant criterion.

Another consideration in the selection of sampling period, completely unrelated to precision, is the advantage of short-term samples in yielding a better definition of the occurrence and magnitude of peak concentrations which is often useful information for diagnostic purposes.

The applicability of equation (25) can be tested experimentally in a given atmosphere by collecting, simultaneously, consecutive sets of samples each set for different time periods. Coefficients of variation of the samples in each set should be calculated and then their reciprocals plotted against the square-roots of the respective sampling periods. Agreement with the equation is signified by a straight line through the data and intersecting the origin.

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