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RADIOACTIVE AEROSOLS

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[1]Disclaimer: The findings and conclusions in this chapter are those of the author and do not necessarily represent the views of the National Institute for Occupational Safety and Health. Mention of company names or products does not constitute endorsement by NIOSH.

28.1 INTRODUCTION

Measurement of radioactive aerosols involves most of the standard tools of aerosol science and technology, as well as a number of specialized techniques that take advantage of

the unique physical properties of radioactive materials. Instruments and techniques for characterizing radioactive aerosols have been described extensively in numerous manuscripts, books, and standards. Detailed review and application of this information, as well as the development of new techniques and applications, continue to occupy the careers of many aerosol scientists and health protection professionals. This chapter provides an overview of the principles, techniques, and applications of measuring radioactive aerosols. Information needed for a basic understanding is presented, and several newer measurement techniques are described.

28.2 RADIATION AND RADIOACTIVE DECAY

Aerosols of radioactive materials have all the physical and chemical characteristics of nonradioactive aerosols. They can range from ultrafine metal fumes to large liquid droplets. Their physical characteristics have the usual influence on their aerodynamic behavior and on the choice of measurement technique. Their chemical characteristics have a similarly important influence on their biological behavior and on the technique that will be used to confirm their chemical form after collection. It is their radioactive properties that can make them both easier to detect, yet, in many cases, more hazardous to handle.

28.2.1 Types of Radiation

Three major types of radiation are of concern in studies of airborne radioactive materials: alpha, beta, and gamma radiation (see Fig. 28-1). Neutrons and positrons are also of concern in some special circumstances. Details of the origin and characteristics of all types of radiation as well as the radioactive decay schemes for radionuclides are presented in references such as the *Table of Isotopes* (Firestone 1996), the *Handbook of Health Physics and Radiological Health* (Schleien et al. 1998), the *Nuclides and Isotopes Chart of the Nuclides* (Baum et al. 2003), and *Radioactive Air Sampling Methods* (Maiello and Hoover 2011).

28.2.1.1 Alpha Radiation Alpha radiation is the least penetrating, but most highly ionizing type of radiation. Alpha particles consist of two neutrons and two protons, carry two positive charges, are identical to a helium nucleus, and are created spontaneously during the radioactive decay of high atomic number elements such as radium, uranium, or plutonium. Alpha-emitting radionuclides can be identified by the characteristic energy of their emissions. They are of special concern when they are inhaled and deposited in the respiratory tract. If inhaled in large quantities, damage to cells in the lung or in other organs to which the material is translocated can cause short-term health effects such as inflammation and subsequent fibrosis and long-term health
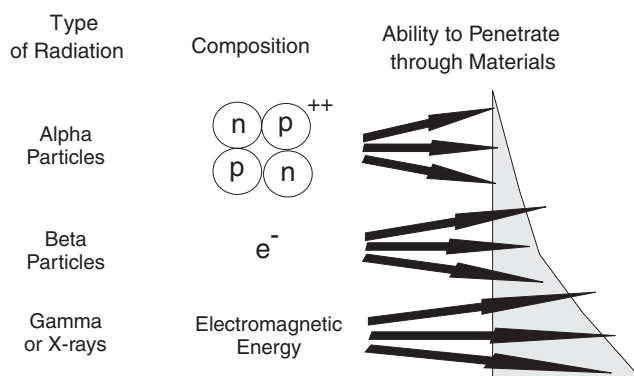

Figure 28-1 Characteristics of the three major forms of radiation of concern for measurements of radioactive aerosols: Alpha particles (consisting of two protons and two neutrons), beta particles (with mass and charge equivalent to that of an electron), and gamma radiation (with a range of electromagnetic energies).

effects such as cancer [c.f., Hobbs and McClellan 1986; Agency for Toxic Substances and Disease Registry (ATSDR) 1990, 1999, 2007, 2008].

28.2.1.2 Beta Radiation Beta radiation is more penetrating than alpha radiation and consists of negatively charged particles that are identical to electrons. Beta radiation can penetrate the skin, and, like alpha radiation, is of concern when large amounts of beta-emitting radionuclides are deposited in the body. The energy spectrum of beta emissions covers a broad range up to a characteristic maximum. Iodine-131, cesium-137, and strontium-90 are beta-emitting radionuclides that are of special concern for accidents involving nuclear reactors.

28.2.1.3 Gamma Radiation Gamma radiation and X-rays are penetrating quanta of electromagnetic energy. They are not charged particles, but they can cause ionization in materials and biological damage in tissues. Gamma radiation originates in the nucleus of atoms during radioactive decay. Gamma radiation is emitted at discrete, characteristic energies for each radionuclide. Many alpha- and beta-emitting radionuclides also emit gamma radiation. X-rays originate in disruptions of electrons from their orbits around the nucleus. X-rays can also arise as a secondary form of radiation called *bremsstrahlung* (braking radiation). Bremsstrahlung is created when the path of a negatively charged beta particle is altered by the coulombic attraction to positively charged nuclei of the absorbing material. This alteration of direction causes a radial acceleration of the beta particle, which is accompanied, in accordance with classical theory, by a loss of electromagnetic energy at a rate proportional to the energy of the beta particle and the square of the atomic number (Z) of the absorbing material (e.g., Cember and Johnson 2009).

28.2.2   Half-Life for Radioactive Decay

The rate at which a radioactive material decays is described by its half-life: the time for half of the atoms present to undergo a spontaneous nuclear transformation. Half-life is an important consideration in determining how to collect, handle, and quantify samples of a radioactive material. For example, samples of short-lived materials may need to be analyzed immediately after collection, or even in real time. Conversely, samples of long-lived radionuclides may remain radioactive for years, centuries, or even millennia. Experimental determination of half-life is also one way of helping to identify a given radionuclide.

The equation describing radioactive decay is

$$A(t) = A_0 e^{-0.693 t / t_{1/2}} \qquad \text{(Eq. 28-1)}$$

where $t_{1/2}$ is the decay half-life for the radionuclide (in time units), t is the elapsed time (in the same time units as $t_{1/2}$), e is the base of the Naperian logarithm system (2.718), 0.693 is the natural logarithm of 2, A_0 is the activity of the sample at time $t = 0$ (in disintegrations per unit time), and $A(t)$ is the activity of the sample after an elapsed time of t.

Another way of writing the relationship involves the decay constant, λ, where

$$\lambda = \frac{0.693}{t_{1/2}}, \quad \text{and} \quad A(t) = A_0 e^{-\lambda t} \qquad \text{(Eq. 28-2)}$$

In terms of the number of elapsed half-lives, n, the amount of activity remaining can be determined as

$$A = A_0 (1/2)^n \qquad \text{(Eq. 28-3)}$$

Thus, after two half-lives, only one-fourth (25%) of the original activity is present, and after seven half-lives only 1/128 (0.8%) of the original activity remains.

28.2.3   Specific Activity

The specific activity of a radioactive material is the rate of decay per unit mass. Materials with a short half-life have a high specific activity. The historical unit for activity, the curie (Ci) (2.22×10^{12} disintegrations/min), was defined in terms of the specific activity (S_A) of radium (1 Ci/g). The international system (SI) unit for radioactive decay is the becquerel (Bq), which is 1 disintegration/s. Thus, 1 Ci is equal to 3.7×10^{10} Bq and 1 nanocurie is 37 Bq. Specific activity is of biological concern for radioactive materials in the body because it determines the rate at which energy will be deposited and damage will occur in tissues. Specific activity is of practical concern for aerosol measurement because it determines the amount of radioactivity that will be present in a sample of a given mass, and the mass or number of particles that will be associated with a sample of a given activity.

The dependence of airborne mass concentration, c_m (g/m^3), on activity concentration, c_a (Bq/m^3), involves the specific activity of the material, S_A (Bq/g), in the following way:

$$c_m = \frac{c_a}{S_A} \qquad \text{(Eq. 28-4)}$$

The number of particles per cubic meter, c_n (particles/m^3), required to provide a given activity air concentration, c_a, involves a similar relationship of specific activity, S_A, particle density, ρ (g/m^3), and the volume of the airborne particles. For a simple example in which particles are assumed to be monodisperse in size and spherical in shape (volume = $\pi d_p^3 / 6$, where d_p is the particle diameter in meters), this relationship is

$$c_n = \frac{c_a}{S_A \rho \pi d_p^3 / 6} \qquad \text{(Eq. 28-5)}$$

28.2.4   Sources of Radioactive Aerosols

Radioactive aerosols occur naturally in our environment from a number of origins, including the interaction of cosmic rays with our atmosphere producing radionuclides such as carbon-14 and beryllium-7; from the dispersion of soil particles (especially phosphate fertilizers) containing potassium-40 or uranium and thorium and their radioactive decay products; and from emanation from soil and building materials of the radioactive noble gases of radon-222 (from the uranium decay series), radon-220 (also known as thoron, from the thorium decay series), and radon-219 (also known as actinon, from the actinium series) (c.f., Cohen 2001). Evans (1969) can be consulted for a historically elegant tutorial on the fundamental principles of in-growth and decay of radon progeny.

Radioactive aerosols are also released by combustion of fossil fuels and by cigarette smoking [e.g., National Council on Radiation Protection and Measurements (NCRP) 2009]. The exposure of humans to radioactive aerosols from cigarette smoke occurs because tobacco contains minute amounts of uranium and thorium decay products in its plant matrix and in residual soil and phosphate fertilizer contamination on leaf surfaces. Lead-210, a beta-emitting radionuclide, and polonium-210, an alpha-emitting radionuclide, are taken up naturally from the soil by tobacco plants. Polonium-210, the alpha-emitting decay product of ^{210}Pb is responsible for most of the radioactivity inhaled as a part of cigarette smoke. The concentrations of these radionuclides have been widely reported in many samples of tobacco and cigarettes from around the world (e.g., Schayer et al. 2009). It has been postulated that ^{210}Po in cigarette smoke contributes to lung cancer in smokers (e.g., Martell 1975).

Additional sources of environmental exposures to radioactive aerosols include the nuclear power fuel cycle and associated accidents, and (historically) open-air testing of nuclear weapons (e.g., Eisenbud 1987). Occupational exposures to radioactive aerosols occur in the nuclear power cycle, in production of nuclear weapons, and in a wide variety of other industrial, agricultural, and biomedical applications of radioactivity [c.f., U.S. Nuclear Regulatory Commission (USNRC) 2000]. An example of accidental dispersion of airborne plutonium during failure of a gasket in a glovebox enclosure has been described by Cheng et al. (2004), and generation and dispersion of aerosols from depleted uranium penetrators of armor-piercing munitions has been described by Parkhurst et al. (2005, 2009). The NCRP (2009) summary of overall exposures with estimated total of 6.2 mSv (620 mrem) of the U.S. population to ionizing radiation in 2006 notes that half of the total exposure comes from natural radiation in soil and rocks, radon gas that seeps into homes and other buildings, plus radiation from space and radiation sources that are found naturally within the human body; approximately half of the exposure is from medical uses of ionizing radiation; and only a small contribution of exposure to the U.S. population arises from consumer products and activities, industrial and research uses, and occupational tasks.

28.2.5   Inhalation Exposure Limits

Statutory limitations on exposures of humans to radioactive materials are provided in regulations such as the USNRC *Standards for Protection against Radiation* (USNRC 2006) and the U.S. Department of Energy regulation on *Occupational Radiation Protection* (USDOE 2007). Specific activity and biological behavior of materials are considered in determining acceptable Annual Limits on Intake (ALIs) for workers exposed to radioactive aerosols [see ICRP (1979) and its addendums]. According to the approach described in ICRP Publication 30 (ICRP 1979), the ALI is the annual intake of a given radionuclide that would result in either a committed effective dose equivalent of 50 mSv (5 rem) or a committed dose equivalent to an organ or tissue of 500 mSv (50 rem). The more recent recommendations described in ICRP Publication 60 (ICRP 1990) do not consider deterministic (organ/tissue) effects in the determination of an ALI, and the reference dose used is 20 mSv (2 rem).

A related concept is the Derived Air Concentration (DAC). A DAC is the calculated air concentration to which a worker could be exposed 8 h/day, 5 days/week, for 50 weeks (an entire work year of 2000 h), without exceeding the ALI for the radionuclide. The worker is assumed to breathe at the rate of 20 L/min, as given for the ICRP Reference Man (ICRP 1975). Thus, the DAC is the ALI divided by the volume of air inhaled during the working year (2400 m^3). Cumulative exposure is expressed in terms of DAC-hours

(e.g., exposure to an air concentration of 1 DAC over a period of 8 h would result in an exposure of 8 DAC-hours).

Note that although the recently issued USDOE regulation *Occupational Radiation Protection* adopted the newer dosimetric models of ICRP Publication 60, the regulation still considered both stochastic and deterministic effects in deriving the ALI and DAC values based on either a committed effective dose of 50 mSv (5 rem) or a committed equivalent dose to an organ or tissue of 500 mSv (50 rem), whichever is more restrictive.

Although the exposure units of ALI and DAC are used for individual radionuclides such as plutonium and uranium in the workplace, an alternate exposure unit known as the Working Level (WL) has historically been used for exposure to radon and its decay products in the uranium mining and milling industries. Exposure to the three predominant isotopes of radon includes exposure to radon decay products involving a variety of emissions of alpha, beta, and gamma radiation. The premise of the WL is that lung cancer is the concern for human health effects following long-term exposure to radon and radon progeny, and that the radiobiological risk for lung cancer arises from inhalation exposure to the high-energy alpha emissions from radon decay products. In other words, the amount of biological damage from any beta and gamma emissions and from radioactive decay in the respiratory tract of radon gas itself is small compared to radiation damage from the alpha decay in the respiratory tract of the accumulated polonium, bismuth, and lead decay products of radon.

By definition, a concentration of 1 WL means the concentration of any combination of short-lived radon decay products in 1 liter of air that will release 1.3×10^5 million electron volts (MeV) of alpha energy during decay. As noted in the toxicological profile for radon (ATSDR 2008), when radon is in equilibrium with its progeny (i.e., when each of the short-lived radon progeny is present at the same activity concentration in air as ^{222}Rn), each pCi of radon in air will give rise to (almost precisely) 0.01 WL (USEPA 2003a). The concept of equilibrium equivalent concentration (EEC) is used to account for the fact that the concentration of decay products are not always in equilibrium with the concentration of radon. A lack of equilibrium is frequently the case when exposure occurs to radon gas that has just been released from soil, water, or building materials into fresh or recently filtered air. If an equilibrium factor (F) is known, then an EEC can be calculated. For example, based on an assumed equilibrium of 40% ($F = 0.4$) between radon and radon progeny in the home [National Academy of Sciences (NAS) 1999], each pCi of radon in air will give rise to approximately 0.004 WL of radon progeny.

Cumulative exposure for worker exposure to radon decay products in mines is expressed in terms of the Working Level Month (WLM), where exposure at a concentration of 1 WL for a period of 1 working month (WM) results in a cumulative

exposure of 1 WLM. A working month is assumed to be 170 h. The Mine Safety and Health Administration (MSHA 1971) regulation on *Safety and Health Standards Underground Metal and Nonmetal Mines* has established a radon exposure limit of 4 WLM in any calendar year. The National Institute for Occupational Safety and Health (NIOSH 1987) criteria document for radon in underground mines has recommended that exposure to radon progeny in underground mines shall not exceed 1 WLM per year.

As noted in the ATSDR toxicological profile for radon (ATSDR 2008), measurements in homes are typically made for radon gas and are expressed in Bq/m^3 or pCi/L of air. It is instructive to calculate the number of WLM received annually by a person living at a residence in which the radon concentration is 1 pCi/L. To convert from residential exposures expressed in pCi/L, it is considered that 70% of a person's time is spent indoors and that 1 pCi/L of radon in the indoor air is equivalent to 0.004 WL of radon progeny (NAS 1999; USEPA 2003a). Given that there are a total of 8,760 h in a year compared to 170 h in an assumed WLM, a person living for 1 year in a residence at a concentration of 1 pCi/L will receive a cumulative exposure of 0.004 × 8760/170 = 0.114 WLM.

28.2.6 Considerations of Particle Size and Solubility

Dorrian and Bailey (1995) have summarized the particle size distribution of radioactive aerosols that have been measured in a wide range of industrial operations. The typical particle size distribution had an activity median aerodynamic diameter (AMAD) of 5 μm with a geometric standard deviation of 2.5, although smaller size distributions were observed in operations involving high temperatures or fumes, and larger particle size distributions were observed in operations such as coarse powder handling. The typical size distribution values reported by Dorrian and Bailey have now been accepted as the default values for use in ICRP Publication 66 on the new *Human Respiratory Tract Model for Radiological Protection* (ICRP 1994a). An associated guide

for application of the model has also been provided (ICRP 2003). The previous default assumption for aerosol particle size in the workplace had been an AMAD of 1 μm (ICRP 1979). The 1-μm AMAD is still the default value for exposure of members of the public to radioactive aerosols in the environment (ICRP 1994a). The recommendations of the ICRP are internationally accepted as a coherent and consistent approach to radiation protection. An alternate model developed by the NCRP (1997) focuses on fundamental considerations of human respiratory tract structure and function in deriving an alternate mathematical model to describe the deposition, clearance, and dosimetry of inhaled radioactive substances.

Workplace data on particle size distribution and solubility properties can be used in the ICRP model to adjust DAC values for specific situations. The characteristics of aerosols from a wide range of activities such as powder handling, spills, and fires have also been summarized in a handbook on airborne release fractions/rates and respirable fractions for nonreactor nuclear facilities (USDOE 1994).

Neither breathing-zone nor area sampling data should be used to assign radiation dose to workers except in cases where no bioassay method is available and where the sampling method can reasonably be expected to provide a representative sample of the airborne radioactivity (USDOE 1998). Concerns for particle solubility are generally high because of the influence it has on the biological behavior of inhaled material, and because knowledge of solubility is needed to correctly apply biokinetic models and interpret bioassay information from urine, fecal, or blood samples obtained from workers. Other measurement techniques, such as *in vivo* monitoring (whole-body or lung counting) and radiation monitoring for hand and foot contamination or for contamination of workplace surfaces, are also part of a total health protection program. They often signal increased airborne concentrations before worker exposures become excessive.

Particle number concentrations for several materials at their DAC are illustrated in Table 28-1 as a function of

TABLE 28-1 Number of Particles per Cubic Meter as a Function of Monodisperse Physical Particle Size for Selected Toxic Materials at Their Derived Air Concentration (DAC)[a]

Particle Diameter (μm)	$^{238}PuO_2$	$^{239}PuO_2$	Enriched Uranium Oxide	Beryllium Metal
10	0.00007	0.02	54	1910
5	0.0005	0.15	433	15,279
3	0.002	0.7	2007	70,736
1	0.07	19	54,180	1,909,859
0.5	0.5	150	433,443	15,278,875

[a]Note the following: Insoluble ^{238}Pu has a specific activity of 6.44×10^{11} Bq/g and a DAC of 0.2 Bq/m^3. Insoluble ^{239}Pu has a specific activity of 2.26×10^9 Bq/g and a DAC of 0.2 Bq/m^3. For 93% enriched uranium, the specific activity is 2.35×10^6 Bq/g [dominated by the activity contribution from ^{234}U, which is present at 1% by mass (Hoover et al. 1998)] and the DAC is 0.6 Bq/m^3. The effective density of beryllium metal aerosol particles with a slight oxide coating is 2 g/cm^3 (Hoover et al. 1989) and the Occupational Safety and Health Administration occupational exposure limit for beryllium is 2 $μg/m^3$.

particle size. These materials include three that are radioactive (^{238}PuO$_2$, ^{239}PuO$_2$, and enriched uranium) and one that is nonradioactive (beryllium, a toxic metal of concern in many nuclear facilities). This example demonstrates that the number of particles of concern for different radionuclides covers a broad range. Problems with correlations between air sampling results and bioassay can be especially severe for high-specific-activity materials such as ^{238}Pu because a small number of particles can be significant from a dose standpoint (Scott et al. 1997; Scott and Fencl 1999). Stochastic concerns for whether or not an individual particle was inhaled do not exist for radionuclides with lower specific activity (for example, tritiated particulate materials), where the aerosol cloud can be expected to be more homogenous at concentrations of concern.

28.3   RADIATION DETECTION

Radiation can be detected in a number of ways. Most of them depend on the ionization process that occurs when gamma rays or charged particles pass through a gas, liquid, or solid and form ion pairs by disrupting atoms and electrons in the material. Radiation detection methods can be used as direct tools for quantifying radioactive aerosols and as support tools for the safe handling of radioactive materials. Measurement of alpha-emitting radionuclides generally requires direct, or nearly direct, contact between the alpha-emitting material and the detection device. Radiochemical preparation or separation of the collected sample may also be required. Proximity requirements are less severe for the more-penetrating beta and gamma forms of radiation. Investigators must have a thorough understanding of the decay schemes of the radionuclides of interest. In many cases the radioactive emissions of progeny radionuclides are of greater biological concern than the emissions of the parent radionuclide. For example, although strontium-90 has a relatively long half-life (28.8 years) and emits beta radiation at a relatively low energy (0.54 MeV), its progeny radionuclide yttrium-90 has a short half-life (64.2 h) and is a high-energy beta emitter (2.28 MeV). An overview of the measuring processes for all three types of radiation is given below. Additional details can be found in resources such as Evans (1955), Mercer (1973), Eicholz and Poston (1979), NCRP (1978a,b), Turner et al. (1988), Turner (1996), Shapiro (1981), Knoll (2000), Cohen and Heikkinen (2001), Schery (2001), Ruzer (2005), Ahmed (2007), Papastefanou (2007), Stabin (2008), the chapter on health physics instrumentation in Cember and Johnson (2009), Maiello and Hoover (2011), and the section on radiation detection principles in the *Operational Health Physics Training* manual (USDOE 1988). Note also that analytical chemistry methods such as mass spectrometry can be used to detect radioactive materials on an atom-by-atom basis, which is useful when isotopic composition can

help identify the origin or source of release of a radioactive material (e.g., USEPA 2006).

28.3.1   Scintillation Counting

Phosphors are materials that absorb energy during the ionization process and re-emit a fraction of it as light flashes (scintillations). Light emission occurs when electrons elevated to a higher energy state make the transition back to the ground state. Scintillation bursts can be detected with a photodiode or photomultiplier device and counted to determine the number of charged particles or gamma rays that passed through the material. The intensity and duration of the scintillation can also be analyzed to determine the energy of the radiation being detected.

A number of different phosphors are suitable for radiation detection. For gamma or low-level beta detection, the most notable are solid inorganic scintillators, such as thallium-doped sodium iodide [NaI(Tl)], cesium iodide [CsI(Tl)], or potassium iodide [KI(Tl)]. The thallium activator serves as an impurity in the crystal structure and enhances the probability that the energy absorbed in the crystal is converted to visible light. Zinc sulfide (usually doped with silver), ZnS(Ag), is an excellent detector for alpha radiation; it is also sensitive to beta radiation and photons. The most commonly used self-luminescent exit signs use ZnS as the scintillator with tritium as the source of photons. Scintillations from the interactions of alpha particles with ZnS are proportional in intensity to the alpha energy and are bright enough to be viewed with the human eye in a darkened room. ZnS must be used as a thin layer (usually a fine crystalline coating on a clear plastic film) because it has a low transmission factor for visible light. Plastic or liquid scintillators such as *trans*-stilbene are also available. Samples containing radioactive materials can be dissolved in a scintillation "cocktail" involving a solvent such as toluene or the more environmentally safe diisopropylnaphthalene to improve the contact between the radionuclides being detected and the phosphors (L'Annunziata 2003). There is a wide range of commercially available cocktail media, many with optimized features for dissolving the test material or functioning at special conditions of pH or salt content.

28.3.2   Ionization Chamber Devices

Gas proportional counters and other ionization chamber devices can detect the number and, in some cases, the energy of radioactive emissions. When gamma rays or charged particles pass through the chamber, ion pairs are formed in the gas. These bursts of ion pairs are drawn by an electric field to a charged sensor, and the number and energy of the emissions can be recorded. Counting efficiency, fidelity of energy determination, and counting rate limits are available for standard ionization devices.

28.3.3   Solid-State Detection

More recently, solid-state detectors have been developed that use special layers of semiconducting materials to provide a site for ion-pair production. These high-efficiency devices provide excellent energy resolution and can be used with a multichannel analyzer to identify the energy spectrum of the detected radiation. In detectors such as high-purity germanium (HPGe) or cadmium telluride doped with a small amount of zinc or chlorine, the detection region extends far enough into the surface that they are highly effective for gamma rays. Ion-implant silicon detectors and surface-barrier detectors have a thin detection layer that is especially useful for alpha and beta radiation detection. When these detectors are used for alpha spectroscopy, the energy resolution is highest when the alpha-emitting material is placed directly on the detector surface, or when the air in the gap between the alpha source and the detector is evacuated. New developments in surface coatings for solid-state detectors are making them highly resistant to damage from liquids, acids, bases, or abrasion.

28.3.4   Autoradiography, Track-Etch, and Other Detection Systems

Radiation-induced ionization can cause chemical changes in materials like photographic film. The film can be developed to reveal the location of the damage tracks. This process is known as *autoradiography*; the radioactive material provides its own exposure of the film. Solid-state track recorders can also be made from materials such as polycarbonate or cellulose nitrate. Following exposure to radiation, the surface of the material can be chemically etched to reveal the location, length, and diameter of the damage tracks. Other detection systems use materials that respond to ionizing radiation by undergoing optical density changes, radiophotoluminescence, thermoluminescence, or conductivity changes (e.g., Chapter 13 in USDOE 1988).

28.3.5   Calibration Considerations

Calibration is a critical part of the detection process. Background samples are analyzed, followed by counting of calibration standards of known activity. The normal calibration process involves the use of sealed sources or electroplated sources with radioactivity levels that are traceable to the U.S. National Institute of Standards and Technology (NIST). Quantities and energies of radioactivity in the calibration sources are generally chosen to correspond to those expected in the samples to be analyzed. This allows appropriate corrections to be made for nonlinear phenomena such as instrument dead time (the inability of a system to detect a new event until it has finished reporting or recovering from a previous event). At high count rates (several million counts per minute or more, depending on the instrument), effects such as dead time can be substantial, but at very low count rates such errors are negligible. When instruments are to be used at very low count rates, higher activity standards are used to ensure that statistically valid counting efficiencies are determined for the instruments. Note that detection uncertainty is generally calculated from Poisson statistics as being proportional to the square root of the number of events detected. Thus, the relative uncertainty is high for low activity sources or for short counting times. The influence of background interference must also be taken into account. For errors of less than 1%, net counts of 10,000 or more are generally required (Price 1965). [The 1% value arises from the fact that Poisson uncertainty for 10,000 counts is equal to 100 (the square root of 10,000) and 100 is 1% of 10,000.] Additional information and guidance on calibration of air sampling instrumentation for airborne radioactivity can be found in Hickey et al. (1991), USNRC (1992), and American National Standards Institute (ANSI 2009).

28.4   OBJECTIVES FOR MEASURING RADIOACTIVE AEROSOLS

As is true for all air sampling and air monitoring, the selection of methods for measuring radioactive aerosols should be determined by the underlying objectives of the sampling effort. "What information is really needed?" "What conditions or interferences will be encountered when the measurements must be made?" "How frequently will the measurements need to be made?" "Who will be assigned to make the measurements?" "What level of training and experience will they have, or need to have?" "What data quality requirements must be met?" and "What other similar questions need to be answered?" This concept seems trivial, but it must be emphasized so that needed data will not be missed, and time and fiscal resources will not be wasted.

As shown in Figure 28-2, sampling objectives can be viewed as a collection of building blocks. Selection of the appropriate objective or objectives can help ensure that airborne materials are properly and successfully assessed and that measurement results are meaningful. Although interrelated, the categories of objectives presented here are sufficiently distinct to warrant separate attention.

An initial and overarching sampling objective is *basic aerosol characterization* to understand the physicochemical and biological properties of aerosols that may be encountered. The understanding gained from this basic characterization step is then used to guide the need for, design of, and execution of sampling initiatives for *worker health protection* to ensure that worker exposures are within allowed limits and As Low As Reasonably Achievable (ALARA); *environmental monitoring* to ensure that environmental releases of
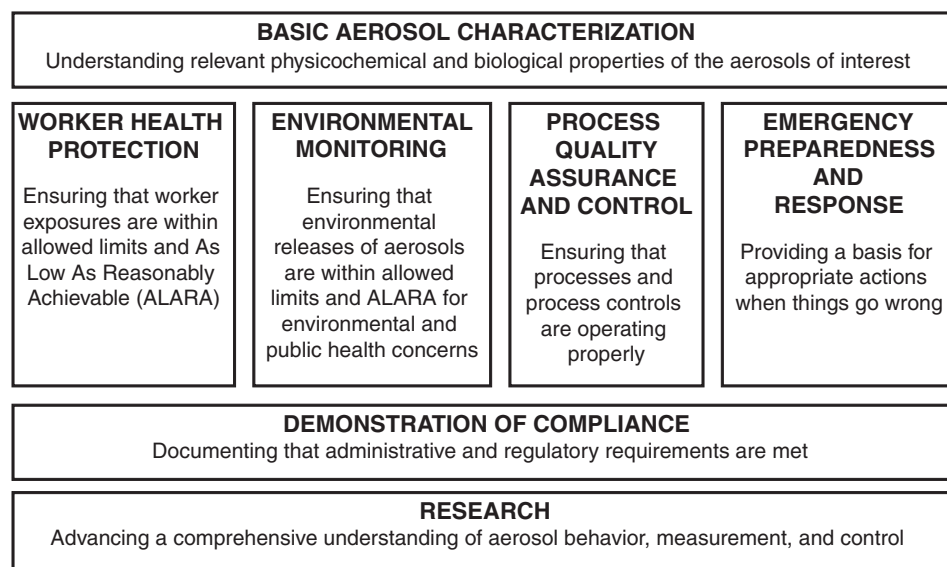
BASIC AEROSOL CHARACTERIZATION Understanding relevant physicochemical and biological properties of the aerosols of interest			
WORKER HEALTH PROTECTION Ensuring that worker exposures are within allowed limits and As Low As Reasonably Achievable (ALARA)	**ENVIRONMENTAL MONITORING** Ensuring that environmental releases of aerosols are within allowed limits and ALARA for environmental and public health concerns	**PROCESS QUALITY ASSURANCE AND CONTROL** Ensuring that processes and process controls are operating properly	**EMERGENCY PREPAREDNESS AND RESPONSE** Providing a basis for appropriate actions when things go wrong
DEMONSTRATION OF COMPLIANCE Documenting that administrative and regulatory requirements are met			
RESEARCH Advancing a comprehensive understanding of aerosol behavior, measurement, and control			

Figure 28-2   Illustration of seven major objectives for sampling radioactive aerosols. The objectives are not mutually exclusive, and other special objectives may also exist.

aerosols are within allowed limits and ALARA for environmental and public health concerns; *process quality assurance and control* to ensure that processes and process controls are working properly; and *emergency preparedness and response* to provide a basis for appropriate actions when things go wrong. Because the Clean Air Act sets limits on allowable releases of radioactive materials to the environment (USEPA 2003b), effluent monitoring or stack sampling is required. This must be done routinely to ensure that environmental releases are within limits and ALARA. Measurements are often made both on site and off site. Because concentration limits for environmental releases are generally lower than for the workplace, greater sensitivity is usually required. This is achieved by using higher sampling flow rates, sampling for longer periods of time, or using more sensitive analytical techniques. Good emergency response involves graded levels of reaction, depending on the severity of any releases. This implies a greater reliance on real-time or near-real-time information than is necessary during normal operations. In addition, instruments may need to remain operational and provide usable results at aerosol concentrations much higher than are seen in routine process, health protection, or environmental monitoring. It is sometimes necessary to deploy portable or mobile instrumentation. Instruments may also need more frequent cleaning, calibration, or replacement.

Because sampling that is conducted on a voluntary or exploratory basis for worker health protection, environmental protection, process quality assurance and control, or emergency planning and response may not necessarily meet formal administrative or regulatory requirements for demonstration of compliance, a distinct objective is identified for *demonstration of compliance* to document that administrative and regulatory requirements are met. Throughout all the measuring regimes, it is necessary to demonstrate that the instruments are working properly and to document actual releases of airborne radionuclides in comparison to statutory release limits. In a sense, this effort is a subset of all other measuring objectives. Once the operator is convinced of what has occurred, the regulators and other interested parties must be convinced.

And, finally, as a fundamental underpinning of all the sampling approaches, theories, methods, practice, and applications, a supporting and unifying objective is included for *research* to advance a comprehensive understanding of the behavior, measurement, and control of aerosols.

These objectives are not mutually exclusive, and other special objectives may also exist.

28.5   APPLICATION OF STANDARD MEASUREMENT TECHNIQUES

Nearly all standard aerosol sampling techniques can be applied to the measurement of radioactive aerosols. Some limitations are related to the amount of material that may be significant or available. For example, the mass amounts of concern for radionuclides may be below the limits of detection of piezoelectric mass monitoring systems or optical monitoring devices. On the other hand, radioactivity levels associated with particles collected for electron microscopy may require use of designated and restricted equipment. Considerations such as these are discussed below for a number of standard measuring techniques.

28.5.1 Optical Particle Counting

Optical particle counters (OPCs) have not been widely used for radioactive aerosols, but there are circumstances under which they might provide useful information to help protect workers from exposures to radioactive aerosols, especially to warn of unusual particle releases (See Chapter 13 for more details on OPCs). Optical particle counters have two main advantages: they sample airstreams continuously, and they provide real-time information. Their main disadvantages are that radioactive particles cannot be differentiated from nonradioactive particles on the basis of light-scattering behavior, and that light scattering generally provides an estimate of the physical size distribution of the observed particles, rather than an estimate of the aerodynamic size distribution, which is more relevant for assessing inhalation risks. A number of commercially available instruments are being used to detect nonradioactive aerosols in cleanrooms, to monitor work area dust levels in industries such as mining and textiles, and to provide quality control monitoring for processes such as paint pigment preparation that fabricate or use fine particles. Optical monitoring has also been used in systems for generating and characterizing inhalation exposure atmospheres of radioactive aerosols (e.g., Hoover et al. 1988a). Issues related to their application for radioactive aerosols include: (1) level of detection compared to allowed air concentrations for radioactive aerosols; (2) level of detection compared to background levels of nonradioactive aerosols in the workplace; (3) aerosol characterization requirements to determine relationships between radioactive and nonradioactive aerosols in the workplace; (4) calibration requirements to quantify instrument response to specific aerosols; and (5) health protection management strategies for using optical monitoring information in a total program for workplace control and worker protection.

Figure 28-3 illustrates a practical application of an evaluation scheme (Hoover and Newton 1991a) for applying an optical particle monitor to the three radioactive materials ($^{238}PuO_2$, $^{239}PuO_2$, and enriched uranium) and one nonradioactive material (beryllium) presented earlier in Table 28-1. For convenience, the abscissa units of Figure 28-3 are DAC. That equalizes the scale for all radioactive materials and also allows treatment of nonradioactive materials. The particle size in the Figure 28-3 example is assumed to be monodisperse 3-μm-diameter spheres. Optical particle counters typically give information as "number greater than a given size." The saturation limit is assumed to be $10^8/m^3$. The lower limit of detection depends on the flow rate and sampling interval. For a flow rate of 0.03 m^3/min (nominal 0.1 cfm, typical of small, portable units) and a sampling interval of 1 min, detection of a single particle during the sample interval would correspond to a concentration of 347 particles/m^3. Figure 28-3 indicates that optical particle counting will not be effective for $^{238}PuO_2$ and $^{239}PuO_2$ because
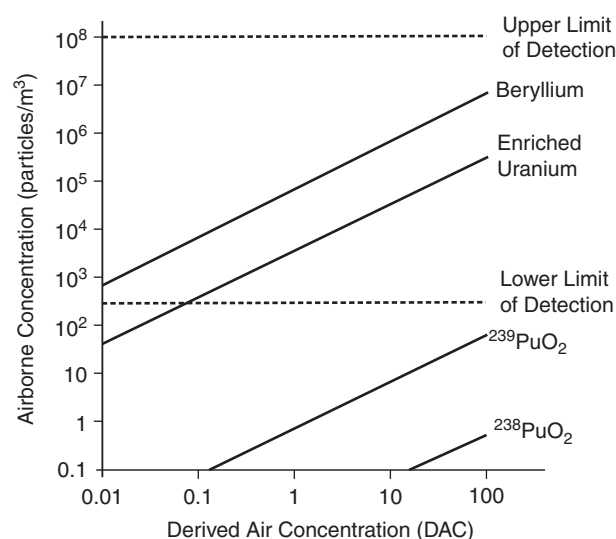

Figure 28-3 Illustration of the evaluation scheme for usefulness of optical particle monitors. Particles are assumed to be monodisperse with diameters of 3 μm. Sampling flow rate is 5×10^{-5} m^3/s [3 L/min].

number concentrations of concern are below normal limits of detection. However, optical particle counting may be useful for enriched uranium and beryllium metal (assuming that background levels of other dusts are not excessive).

28.5.2 Particle Collection for Microscopy

Collection of radioactive particles for morphological examination by transmission or scanning electron microscopy can easily be done using standard instruments such as the point-to-plane electrostatic precipitator (Morrow and Mercer 1964). A small hood is normally sufficient for sample handling. The airflow rate through the hood opening should be maintained at the lowest effective level (approximately 75 linear feet per minute) to avoid problems in handling the small, fragile electron microscope grids. Standard Formvar®-coated copper grids can be used for transmission electron microscopy. Degradation of the Formvar by radiation damage is usually only a problem for high-specific-activity radionuclides such as ^{238}Pu (half-life = 87.7 years).

Care should always be taken to use contamination control features, such as liquid nitrogen cold fingers, to minimize the spread of contamination within the microscope. High-specific-activity, alpha-emitting radionuclides such as ^{238}Pu are prone to migration from the collection grid by radioactive decay-induced recoil and fragmentation (spallation) of particles. The need for respiratory protection and radiation monitoring should be considered before servicing any microscope used for radioactive materials. Chapter 10 provides a comprehensive overview of different microscopy and microprobe analysis techniques.

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28.5.3 Filtration

Filtration is the most widely used method for collecting samples of radioactive aerosols (see Chapter 7 for conventional filter-based sample collection techniques). The methods and equipment range from high volume samplers (sampling rates up to about $60 \, \mathrm{m}^3/\mathrm{h}$) for environmental or short-term workplace sampling, to low volume miniature lapel samplers (1 L/min or less) for collecting aerosols in the breathing zone of individual workers (USDOE 1988). Low pressure drop cellulose filters are commonly used, and samples can be easily reduced to ash or dissolved for analysis using chemical or radiochemical procedures.

Concerns for penetration of particles into the filter matrix are a function of the type of filter, the type of radiation, and the radiation counting method being used. Membrane filters with their superior front-surface collecting characteristics are preferred over fiber-type filters when alpha particle spectroscopy is applied. Shielding by the filter media is seldom a concern for detection of gamma radiation. Although energy degradation concerns are greatest for alpha-emitting radionuclides, Hoover and Newton (1993) reported that even glass microfiber filters such as the *GEL*[2] A/E glasscollect particles near enough to the filter surface that radioactivity counting results from the ZnS method are as accurate as radiochemical results.

Long-term storage of filter samples for archival purposes is not always feasible for high-specific-activity, alpha-emitting radionuclides such as ^{238}Pu. Radiation damage to the filter, packaging tape, or plastic container may allow release of radioactivity. Care should be taken when handling samples that have aged. Care should also be taken during sample collection. Portable aerosol sampling systems may be used to provide local ventilation and filtration around the sampling instruments (e.g., Hoover et al. 1983). Ultraviolet- and chemical-resistant glove materials such as Hypalon® and specially designed modular glovebox links may be used to minimize the spread of contamination and exposures of workers (Hoover et al. 1999). Additional information for control and safe handling of radioactive materials can be found in Maiello and Hoover (2011).

28.5.4 Inertial Sampling

Inertial sampling using cascade impactors, spiral duct centrifuges, and cyclones has been the major approach for characterizing the aerodynamic particle size distribution of radioactive aerosols. See Chapter 8 for conventional inertial sampling techniques. A number of specialized versions of these instruments have been developed specifically for use with radioactive aerosols (e.g., Mercer et al. 1970; Kotrappa

[2]See Appendix I for a complete list of manufacturers' addresses indexed to three-letter codes.

and Light 1972). Special requirements for instruments used in handling radionuclides generally include being compact and easy to assemble and disassemble while wearing protective gloves in a confined space such as a glovebox enclosure. They also need to be easy to clean. Low sample collection rates are usually adequate for collection of small sample masses. Analytical methods for radioactive counting of collected samples are straightforward. The spiral duct centrifuge has been used to estimate the density or shape factor of individual particles. This works equally well for radioactive and nonradioactive particles. Because the aerodynamic diameter associated with each particle deposition location is known, electron microscopy can be used to determine the physical size and shape of particles found at those locations, and density or shape factor can be calculated (Stöber and Flachsbart 1969).

Real-time inertial techniques, such as time-of-flight measurements of particles accelerated through a nozzle, are also useful for radioactive aerosols. This assumes a willingness to purchase dedicated instruments for use with radioactive aerosols, because decontamination of equipment for return to unrestricted use is not always easy.

28.5.5 Measurement of Electrical Properties

Measurement of aerosol electrostatic charge distribution can be done using a standard aerosol charge spectrometer (Yeh et al. 1976; see Chapter 15). The theory of Yeh et al. (1976, 1978) predicts that self-charging due to alpha or beta emission in radioactive aerosols will occur in addition to friction charging due to comminution. Even when aerosols are created by highly charging processes like grinding, radioactive decay processes such as alpha or beta decay may quickly result in a charge distribution that is near Boltzmann equilibrium. Measurements of particle charge distribution on a plutonium-uranium aerosol obtained by Yeh et al. (1978) with and without use of a krypton-85 discharge unit were identical. At high alpha radioactivity concentrations ($>25 \, \mathrm{nCi/L}$), it is likely that sufficient ion pairs are present to reduce the charge on the aerosols to near Boltzmann equilibrium.

At lower radioactivity concentrations, this equilibrium condition may not be reached. Raabe et al. (1978) reported the anomalous results of two cascade impactor samples taken without the use of a ^{85}Kr discharge unit in the blending step of a mixed plutonium-uranium oxide fuel preparation process. The alpha radioactivity concentration at the time those samples were taken was only 1 to 2 nCi/L. Because the AMAD of these samples was larger than observed in samples taken with a discharger, it is likely that anomalous deposition on the upper stages of the impactors occurred as a result of electrostatic charge effects. In the absence of advance information on the radioactivity concentration being sampled, inclusion of an in-line ^{85}Kr, ^{210}Po, or other

type of discharge unit is therefore recommended as a standard procedure, even though it may not be needed to reduce the charge distribution to Boltzmann equilibrium. Fabrication and use of ^{85}Kr discharge units are described by Teague et al. (1978). Krypton-85 discharge units are commercially available (e.g., **TSI**).

Users of ^{85}Kr discharge units should be aware that krypton is a gas that can leak from the metal tube that retains the gas within the discharge unit, and that ^{85}Kr has a radioactive half-life of 10.756 years, which results in a steadily decreasing source strength. Thus, it is prudent to monitor the source strength over time. This can be done by using a Geiger counter to periodically measure and maintain a control chart of the radiation dose rate at the surface of the discharge unit. The measured dose rate will be the result of bremsstrahlung from interaction of the pure beta emissions from ^{85}Kr with the structure of the discharge unit.

28.5.6   Volumetric Grab Samples, Impingers, Cold Traps, and Adsorbers

Sampling techniques such as evacuated volumes, liquid bath impingers, cold traps, and activated charcoal adsorbers work equally well for capturing radioactive and nonradioactive vapors and particles. Standard radioactivity counting techniques can be applied, depending on sample geometry. The Lucas cell (Lucas 1977; NCRP 1988) is a grab-sampling approach in which radioactive particles or gases are drawn into a chamber whose interior walls are coated with a layer of crystalline ZnS(Ag) or other scintillator. Nearly 100% of the alpha particles reaching the scintillator will result in a flash of light. Any flashes of light occurring within the chamber are observed by a detector. This method can be applied to radon gas, radon decay products, or other alpha-emitting radionuclides that can be drawn into the chamber. The half-life of the radionuclides being sampled influences the delay time or cleaning requirements before the cells can be reused.

28.5.7   Analytical Chemical Techniques

Traditional analytical chemistry methods such as infrared spectrometry, flame or furnace atomic absorption spectrometry, energy dispersive X-ray analysis, electron or neutron diffraction, and inductively coupled plasma (with either emission spectroscopy or mass spectroscopy) have sensitivities that are compatible with the small sample sizes usually associated with radioactive aerosols (see Chapter 9 for standard aerosol chemical analysis methods). Sensitivities of various techniques have been compiled and reported (USEPA 2006). Dedicated equipment is usually required for handling radionuclides, and appropriate controls must be applied. Techniques requiring tens or hundreds of milligrams, such as X-ray diffraction, have much more limited application for radioactive aerosols.

28.6   SPECIAL   TECHNIQUES   FOR RADIOACTIVE   AEROSOLS

28.6.1   Detection of Individual Particles by Autoradiography

The interaction of ionizing radiation with photographic film or solid-state nuclear track detectors such as CR-39 (a polycarbonate plastic) creates tracks or material defects that can be photographically developed or chemically etched for examination by scanning electron microscopy or light microscopy. The location, number, and length of the tracks can be used to determine the position and estimate the radioactivity of individual radioactive particles. In one example, Voigts et al. (1986) identified single aerosol particles as the alpha sources from industrial plume samples with particle number concentrations of 2,000 particles/mm^2. Cohen et al. (1980) used cellulose nitrate track-etch film to measure the alpha radioactivity on human autopsy specimens of the bronchial epithelium. In combination with microscopy, autoradiography can be used to determine how cells and organelles are irradiated by particles that are inhaled and deposited in the body. Figure 28-4 shows an example of autoradiography used with a histological slide of lung tissue to determine the microdosimetry of an inhaled uranium-plutonium oxide aerosol.

28.6.2   Measurement of Particle Solubility to Infer Biological Behavior

Particle solubility and biokinetic studies can be done on all classes of aerosol particles, but measurement of dissolved material is especially straightforward for radioactive aerosols (Kanapilly et al. 1973). Samples can be sandwiched between filters and subjected to continuous solvent flow (dynamic systems) or placed sequentially in fresh containers of solvent (static systems). Particles can also be placed in a tube with the solvent and periodically centrifuged to concentrate the particles in the bottom of the tube and to allow sampling of dissolved material from the supernatant. Radioactivity counting provides the very low limits of detection needed for accurate determination of particle dissolution, especially for highly insoluble materials, and radioactive aerosols have played a unique role in many unusual discoveries about aerosol behavior. The high degree of measurement sensitivity was a major factor in the studies of Mewhinney et al. (1987a) that showed a rapid initial release of material whenever particles were reintroduced into a solvent. That work provided an insight into the possible environmental effects of wet and dry weathering on particles released to the biosphere. Current practice involves conducting *in vitro* dissolution studies under conditions that simulate the important biological compartments of the extracellular fluid of the lung, which has near-neutral pH (7.2–7.4) and the intracellular
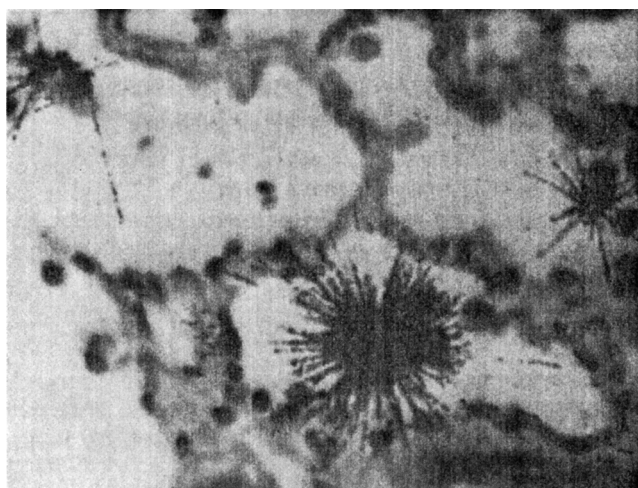
Figure 28-4 Autoradiograph of alpha-emitting particles of varied specific activity in the lung of a rat following inhalation exposure to mixed uranium-plutonium oxide aerosols. Adapted from Mewhinney (1978).

phagolysosomal environment where phagocytized materials are subjected to a more acidic pH (4.5–5) (Ansoborlo et al. 1999). Stefaniak et al. (2005) have developed a simulated phagosomal stimulant fluid that complements the simulated lung fluid of Kanapilly et al. (1973). In addition, Stefaniak et al. (2007), building on the historical model of Mercer (1967) for the dependence of dissolution on particle size and surface area, provide an improved theoretical framework for the use of measured particle properties to evaluate analytical digestion methods for poorly soluble particulate materials.

28.6.3 Density Measurement by Isopycnic Gradient Ultracentrifugation

Isopycnic density gradient ultracentrifugation has been shown to be a useful technique for measuring the density of small quantities (0.1–5 mg) of a variety of particles (Allen and Raabe 1985; Finch et al. 1989). Normal density measurement techniques, such as air or gas pycnometry or liquid displacement, are not suitable for the small sample volumes normally associated with radioactive aerosols. Thallium formate has been the usual heavy-metal solution for this technique, but sodium metatungstate has been shown to be an economical, nontoxic alternative (Hoover et al. 1991). Figure 28-5 illustrates the technique. Particles are added to a centrifuge tube containing the heavy liquid. The tube is subjected to centrifugation to form a gradient of density from top to bottom. Density near the top of the tube normally approaches that of water, and higher density near the bottom of the tube can be higher than 3.0 g/cm^3. Particles move to the location within the tube where their density equals that of the surrounding liquid. Successive samples of known volume are then removed, weighed to confirm the density of the liquid in the sample, and then analyzed by radioactivity counting or other suitable methods to determine the fraction of the particle material in the sample. Refractive index measurements of the liquid samples can also be used to determine the density of the liquid samples, but that requires a dedicated refractometer and is usually more time-consuming than simple weighing. The density gradient ultracentrifugation technique provides information about the density distribution of particles within an aerosol sample.
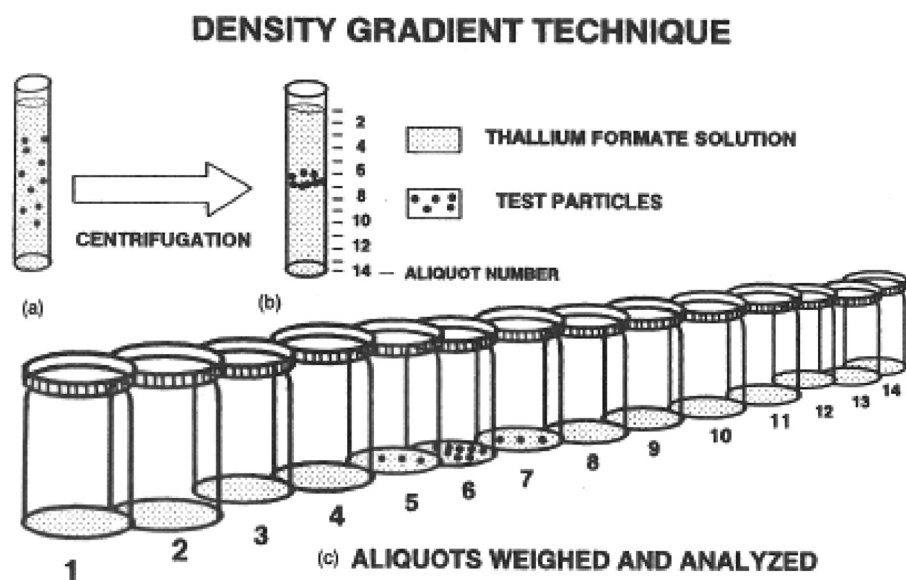

Figure 28-5 Illustration of the density gradient ultracentrifugation technique for determining the density of individual particles. Adapted from Finch et al. (1989).

28.6.4 Surface Area Measurement by Krypton-85 Adsorption

Particle specific surface area (m^2/g) influences the rate of surface phenomena such as dissolution (c.f., Mercer 1967). When adequate sample masses are available (10–50 mg, or more), measurement of specific surface area is reliable and straightforward. The most widely used approach involves the "BET" method (Brunauer, Emmett, and Teller 1938) of calculating nitrogen adsorption onto the surface of a sample of known mass. Other gases such as oxygen, helium, carbon dioxide, krypton, and methane have also been used. A number of commercial instruments are available. Rothenberg et al. (1982, 1987) have focused attention on the special problems of surface area measurement when sample size is less than 10 mg, which is a typical restriction when characterizing radioactive particles. They have described and evaluated a method for adsorbing ^{85}Kr gas onto the sample surface (Rothenberg et al. 1987). Radioactive decay of ^{85}Kr emits a 0.514-MeV gamma ray that can be readily detected by a standard scintillation method such as NaI(Tl). They note that a 1-cm^2 monolayer of ^{85}Kr gas having a specific activity of 10 Ci/g will give approximately 10,000 disintegrations/min that can easily be measured with an uncertainty of less than 1%. The major statistical uncertainty is associated with blank correction for the sample holder. The ^{85}Kr adsorption technique has been successfully applied to the characterization of small samples of mixed uranium and plutonium dioxide particles (Mewhinney et al. 1987b), and the technique can be used for samples as small as 1 mg, with specific surface areas greater than 1 m^2/g. However, the disadvantage of the method rests in the lack of a commercially available instrument, and the significant effort required for a user to set up the method.

28.6.5 Real-Time Monitoring for Airborne Radionuclides

The International Electrotechnical Commission (IEC) has developed standards for radiation protection instrumentation for continuously monitoring radioactivity in gaseous effluents, including general requirements (IEC 2002a) and specific requirements for: (1) radioactive aerosol monitors, including transuranic aerosols; (2) radioactive noble gas monitors; (3) iodine monitors; and (4) tritium monitors (IEC 2002b–e, 2007, 2008). Many instruments for real-time monitoring of airborne radioactivity are commercially available. The typical instrument configuration involves a radiation detector in close proximity to a filter that is collecting aerosols. Radioactivity concentration information can be accessed at the instrument itself, or instruments can be networked to central monitoring stations. Radiation shielding and background correction for external radiation sources are required for beta and gamma radiation detection systems, and a range of background correction, calibration, and geometry considerations come into play for alpha radiation

detection systems. Some of the important considerations for design, calibration, and operation of continuous air monitors (CAMs) for alpha-emitting radionuclides are given below.

28.6.5.1 Mitigation of Interference from Radon Progeny The alpha emissions of naturally occurring radon progeny such as ^{218}Po and ^{212}Bi (with alpha energies of 6.0 MeV and 6.08 MeV, respectively) are similar enough in energy to the alpha emissions of ^{239}Pu (alpha energy 5.2 MeV) and ^{238}Pu (alpha energy 5.5 MeV) to cause interference or false-positive reports of plutonium air concentrations. Early alpha CAM designs did not include spectrometry, but used a single-channel analyzer to detect radioactivity in a plutonium energy region of interest (ROI), and a second analyzer to both detect radon progeny activity in a second energy region and to allow a simple background correction for the counts seen in the plutonium ROI. The correction method was crude and the limit of detection high, but it provided a useful, real-time means for detecting relatively large airborne releases of plutonium.

An alternate approach to handling radon progeny background interference was developed at the USDOE Savannah River Site (Tait 1956; Alexander 1966). It used an impactor jet to deposit aerosol particles directly onto a photomultiplier tube coated with a thin layer of ZnS(Ag). This eliminates most of the radon progeny, which are usually attached to the small particle fraction of ambient aerosol (diameter $<$ 0.3 μm) and are smaller than the effective cutoff diameter of the impactor, and provides a real-time capability for detecting plutonium particles deposited on the collection substrate (Chen et al. 1999). Smaller plutonium particles are also undetected, but most aerosol releases in the workplace have an AMAD of about 5 μm with a geometric standard deviation of about 2.5, and therefore, most of the radioactivity is associated with larger particles (Dorrian and Bailey 1995). Radioactivity counting efficiency for the collected plutonium was approximately 50% (nearly 100% of the emissions occurred in the direction of the ZnS layer) because the light emissions from the ZnS(Ag) are emitted isotropically, and all emissions can be seen with equal likelihood by a photomultiplier tube or photodiode detector. A variation of the Savannah River approach used an impactor jet to deposit the aerosol directly onto the surface of a solid-state detector (Model 8300, ***KUR***). Although detection efficiency for collected material was excellent, and direct detection of the alpha particles causes good separation of the plutonium and radon progeny peaks, particle bounce off the detector surface degraded the collection efficiency of large particles. Hoover and Newton (1993) reported that they found more than 90% of the 10 μm AMAD particles to be lost from collection due to bounce. Particle loss by bounce might be solved by use of a virtual impactor or a particle trap approach. Biswas and Flagan (1988) provide a description of a particle trap.

Advances in microcomputers have enabled a major development effort to include alpha spectroscopy in solid-state

detector CAMs for real-time monitoring of alpha-emitting radionuclides. Again, a solid-state detector is placed just above the surface of a filter onto which alpha-emitting aerosols are drawn, but the detector output goes to an embedded spectrometer in the CAM, rather than to single-channel analyzers. In a batch operation, the air gap between the filter and the detector can be partially evacuated. This reduces attenuation of the alpha particle energies as they pass through the air gap and minimizes any spectral overlap of the plutonium and radon progeny alpha energy peaks. For real-time sampling of the aerosol, however, the region between the filter and the detector cannot be evacuated, and attenuation of the alpha particle energies occurs in the air gap.

The first successful algorithm for subtraction of radon progeny interference in the plutonium energy region was developed by Unruh (1986) at Los Alamos National Laboratory. Four ROIs were established: ROI-1 on the lower energy tail of the ^{218}Po peak, where plutonium is expected; ROI-2 on the higher-energy tail of the ^{218}Po peak; ROI-3 on the lower-energy tail of the ^{214}Po peak; and ROI-4 on the higher-energy tail of the ^{214}Po peak. Basically, the ^{218}Po and ^{214}Po peaks were assumed to be congruent in shape, and the radon progeny activity in the plutonium ROI (ROI-1) was estimated by taking a ratio of the radon progeny counts in the other three regions of interest:

$$(\text{ROI-1}) = \text{k} \cdot (\text{ROI-2}) \cdot (\text{ROI-3})/(\text{ROI-4}) \qquad (\text{Eq. 28-6})$$

This approach was used in the Eberline Alpha 6 (**TFS**) and in the Victoreen Model 758 (Victoreen, Inc., Cleveland, OH). The Radeco Model 452 (SAIC/Radeco, San Diego, CA) used a similar detector and filter arrangement with a triple-window background subtraction algorithm (counts in regions above and below the plutonium region were used to make the background subtraction), or a peak-shape subtraction algorithm that used peak stripping to remove interfering counts from the plutonium ROI. Note that these CAMs based on the region-of-interest approach are still in use in some nuclear facilities, but are no longer commercially available.

The **CAN** Alpha Sentry, **EBE** Alpha 7, Lab Impex SmartCAM (Lab Impex, Poole, Dorset, UK), and the Bladewerx Sabre-series alpha CAMs (Bladewerx, Rio Rancho, NM) use peak-shape algorithms for correction of radon progeny background. The **CAN** Alpha Sentry also includes a fine screen on the aerosol inlet to prevent unattached radon progeny from penetrating to the aerosol collection region of the instrument (McFarland et al. 1992). The **CAN** Harwell iCAM has a moving filter for extended sampling and elimination of interference from previously collected aerosol. A personal sampling version of the Bladewerx CAM is available.

The **MGP** alpha-beta CAM uses a very large detector-to-filter separation distance (more than 1 cm) and a stainless steel baffle assembly (installed between the filter and the detector) to collimate the "beam" of alpha emissions reaching the detector. Although the overall detection efficiency is severely reduced because of the small solid angle of view from the detector to the filter, the collimation results in a very narrow full-width-at-half-maximum (i.e., very good resolution) for the alpha energy peaks. That obviates the need for a correction for radon progeny interference in the plutonium ROI.

Other instruments have used coincidence-counting measurements of beta or gamma emissions from radon progeny to correct for alpha emission interference. However, these indirect measurements are susceptible to errors when the composition or concentration of the radon progeny background is altered.

Performance testing of alpha continuous air monitors is addressed in an international standard promulgated by the IEC (2002b). Test facilities meeting the requirements of that standard have been described by Hoover and Newton (1998) and Grivaud et al. (1998). Concerns relate to particle collection efficiency and stability of the radon correction algorithms over a range of radon progeny concentrations.

28.6.5.2 Filter Requirements for Continuous Alpha Air Monitoring
An active area of work has related to the selection of appropriate filter media for alpha CAMs. The quality of the alpha energy spectrum reported by a solid-state detector system is strongly dependent on the type of filter substrate used for particle collection. Membrane filters have long been accepted as having both the excellent particle collection efficiency and the excellent surface collection characteristics needed for alpha spectroscopy. Lindeken et al. (1964) demonstrated that, compared to submicrometer filters, large-pore membrane filters show no serious sacrifice in collection efficiency until the pore diameters exceed 5 μm. Thus, larger-pore filters, with their lower pressure drop, are preferred. The ANSI N13.1 (1999) standard states that if a filter with a collection efficiency lower than 95% is required to meet the overall sampling objectives, then a correction for efficiency shall be made.

The filter that has been widely recommended by CAM manufacturers is the 5-μm-pore-size, membrane filter (mixed cellulose ester, type SMWP, **MIL**). However, the fragile nature of that filter was found to be unacceptable (Hoover and Newton 1991b, 1992). Particle collection efficiency and gravimetric confirmation of particle mass collection were unreliable because of filter breakage under field conditions.

Results of evaluations to find a more rugged membrane filter that would have a reasonably low pressure drop and excellent surface collection efficiency are summarized in Annex D of ANSI N13.1 (1999). The Versapor®-3000 (an acrylic copolymer on a nonwoven nylon fiber support, **PAL**) has performance very similar to that of the SMWP (**MIL**). The Durapore™ 5-μm-pore-size, polyvinylidene fluoride membrane filter (**MIL**) is acceptable but provides a poorer alpha energy spectrum than the SMWP. The AW-19 filter

(*MIL*) was a reinforced mixed cellulose ester filter that was rugged and provided a better alpha energy spectrum than the SMWP. It was also easy to dissolve for radiochemical analysis. The AW-19 is no longer available, but the RW-19 (*MIL*) provides similar durability to the AW-19 with an alpha energy spectrum similar to the SMWP. Performance of the 3-μm-pore-size Fluoropore™ FSLW filter (*MIL*) was found to be superior to all the others because it has a very low pressure drop and it is an excellent front-surface-collecting filter (Hoover and Newton 1992). It is a polypropylene-backed, polytetrafluoroethylene (PTFE) filter that is extremely rugged and provides excellent alpha energy spectral separation of the collected radionuclides. The comparative quality of radon progeny spectra collected with the Fluoropore FSLW (*MIL*) and SMWP (*MIL*) membrane filters are shown in Figure 28-6. Note the much lower tail of the ^{218}Po peak in the plutonium region of interest (around 5.2 MeV). As a warning, Figure 28-6 also shows the extremely poor spectral quality obtained when a conventional fiber filter such as the *WHA* 41 is used in an alpha CAM. Thus, the use of fiber-type filters must be avoided. These and other factors that affect alpha particle detection in continuous air monitor applications are presented by Moore et al. (1993).

A useful innovation in filtration technique involves mounting each filter in a thin cardboard carrier. The card protects the filter during handling and can be labeled to readily identify the sample. Bar code labeling and computer data logging can be used for both the filter cards and the sampling instruments. This improves chain-of-custody control and facilitates long-term archiving of samples. If samples are to be destructively analyzed instead of archived, they can be easily removed from the cards. The choice of filter for use in the cards depends on the quality of the energy spectrum that is

Figure 28-6   Illustration of the influence of filter type on the quality of the radon progeny energy spectrum in an alpha continuous air monitor. The peak on the left is from ^{212}Bi (6.08 MeV) and ^{218}Po (6.00 MeV). The peak in the middle is from ^{214}Po (7.68 MeV). The peak on the right is from ^{212}Po (8.78 MeV).

required and on compatibility requirements of the filter for the analytical technique to be used.

28.6.5.3   Mitigation of Interference from Airborne Dust
The accumulation of ambient dust on the collection filter of an alpha CAM leads to attenuation of alpha energy, just as the air gap above the filter degrades the alpha energy. Such burial of plutonium leads to underreporting of air concentrations, ranging from 10% to 100% when airborne dust concentrations are greater than 1 mg/m^3 (Hoover et al. 1988b, 1990). Alpha particles from plutonium that is buried by 20 μg/mm^2 of salt on a filter are prevented from reaching the detector. This does not prevent the CAM from responding to large puff releases of radioactivity, but it does raise the limit of detection for slow, continuous releases. Dust concerns are primarily associated with decommissioning activities where metal piping and structures are being cut (see Newton et al. 1987), with environmental restoration activities where soil is being disturbed, and with special activities such as in the underground salt mine environment of the DOE Waste Isolation Pilot Plant near Carlsbad, New Mexico.

28.6.5.4   Efficiency Considerations for Filter/Detector Geometry
Concerns for dust loading make it necessary to consider some trade-offs in filter and detector size. Many CAMs use a 25-mm-diameter detector and a 25-mm-diameter filter. This arrangement has an overall detection efficiency of approximately 20%. Retaining the same detector size, but increasing the diameter of the filter collection surface to 43 mm cuts the detection efficiency in half (down to 10%), but increases the collection surface area by nearly a factor of 4. At first, this appears to be a reasonable trade-off.

However, a closer examination of detection efficiency as a function of filter diameter reveals that material collected at the filter edges contributes very little to overall efficiency. This is because of solid angle considerations that reduce both the efficiency at which alphas are intercepted by the detector, and the energy at which they are detected. With a larger filter, alpha-particle emissions from the filter edge lose a significant amount of energy as they travel to the detector. Alpha particles traveling from one edge of the filter to the opposite edge of the detector have the longest path through air, and thus suffer the greatest energy loss, often enough to remove them from the plutonium ROI. Plutonium collected at the center of the filter is detected in the plutonium ROI at an efficiency of 30%, with little energy degradation. At a diameter of 25 mm, detection efficiency has dropped to 15% but only marginal energy degradation has occurred. At a diameter of 43 mm, only 0.04% of the emitted alpha particles reach the detector and have energies in the plutonium ROI. Thus, the larger filter has marginal utility, especially in the presence of dust, where additional energy degradation will degrade any alpha particles from the filter edge to energies below the plutonium ROI.
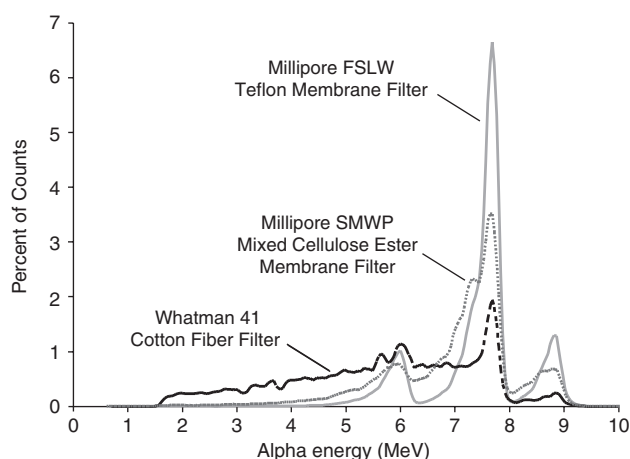
28.6.6   Remote Detection of Radioactive Particles

A technique has been developed at Los Alamos National Laboratory for remotely detecting alpha radioactivity contamination on internal surfaces of equipment or in areas that cannot be directly surveyed by radiation instrumentation (MacArthur and Allander 1991). It uses the fact that ion pairs persist in the air after they are formed by the movement of positively charged alpha particles through air. This persistence was a surprise to many who believed that the recombination of ion pairs was nearly instantaneous. By drawing clean air across a contaminated surface, ion pairs can be carried to an electrometer and detected.

28.6.7   Sampling from Stacks and Ducts

The original version of the ANSI *Guide to Sampling Airborne Radioactive Materials in Nuclear Facilities* (ANSI 1969) has been revised and reissued as the ANSI publication *Sampling and Monitoring for Releases of Airborne Radioactive Substances from the Stacks and Ducts of Nuclear Facilities* (ANSI 1999). Both versions of the standard emphasize the importance of obtaining a representative sample. However, the original standard included faulty assumptions about aerosol mixing in exhaust ducts and faulty technical guidance on the use of multi-point sampling to obtain a representative sample. The revised standard minimizes reliance on simplistic concepts such as isokinetic sampling (considered by many to be broadly misapplied in turbulent sampling conditions) and emphasizes "qualification" of a well-mixed sampling location, which enables the more efficient collection of samples from a single point. Compact and cost-effective mixer designs have been developed to create a well-mixed condition in an exhaust stack (McFarland et al. 1999a,b). The revised standard also contains information about using the more efficient shrouded probes for sample extraction (see, for example, McFarland et al. 1989; Rodgers et al. 1996). Significant improvements have also been made in modeling the deposition of radioactive aerosols in sampling transport lines. Modern codes such as DEPOSITION 4.0 (Anand et al. 1996; Riehl et al. 1996) can be used to predict particle deposition as a function of transport-line configuration. An overview of the methodology for sampling effluent air from stacks and ducts of the nuclear industry is available (McFarland 1998).

28.7   CONCLUSIONS

The measurement of radioactive aerosols is a challenging and specialized subset of aerosol science. The basic physics of radiation and radioactivity provides some unique technical advantages and disadvantages for accomplishing the task. Overall, the advantages outweigh the disadvantages from a measurement perspective. At the same time, a range of political and regulatory obstacles arise from both the rational and irrational health and environmental concerns that surround radioactivity and the use of radioactive materials. Because the health, economic, psychological, and political costs to society can be high, the aerosol measurement tools must be used in a wise and scientific manner when dealing with exposure assessment of radioactive aerosols.

28.9   LIST OF SYMBOLS

A radioactivity (disintegrations per unit time)
c_a radioactivity concentration (Bq/m^3)
c_m mass concentration (g/m^3, mg/m^3, $\mu g/m^3$)
c_n number concentration (particles/m^3)
d_p particle diameter (m, μm)
k radon progeny counting factor; Eq. 28-6
ROI   region of interest, Eq. 28-6
t time
$t_{1/2}$ half life or half time; Eq. 28-1
S_A specific surface area (radioactivity per unit mass)
Z atomic number
λ radioactive decay constant (fraction per unit time)

Subscripts
a   activity
n   number
m   mass
p   particle
0   initial condition

28.10   REFERENCES

Ahmed, S. N. 2007. *Physics and Engineering of Radiation Detection*, New York: Academic.

Alexander, J. M. 1966. A continuous monitor for prompt detection of airborne plutonium. *Health Phys.* 12:553–556.

Allen, M. D. and O. G. Raabe. 1985. Slip correction measurements of spherical solid particles in an improved Millikan apparatus. *Aerosol Sci. Technol.* 4:269–286.

Anand, N. K., A. R. McFarland, V. R. Dileep, and J. D. Riehl. 1996. *Deposition: Software to Calculate Particle Penetration through Aerosol Transport Lines*. NUREG/GR-0006, Washington, DC: U.S. Nuclear Regulatory Commission.

American National Standards Institute (ANSI). 1969. *American National Standard Guide to Sampling Airborne Radioactive Materials in Nuclear Facilities*. ANSI N13.1-1969. New York: ANSI.

American National Standards Institute (ANSI). 1999. *American National Standard for Sampling and Monitoring for Releases of Airborne Radioactive Substances from the Stacks and Ducts of Nuclear Facilities*. ANSI N13.1-1999. New York: ANSI.

American National Standards Institute (ANSI). 2009. *American National Standard on Radiation Protection Instrumentation Test and Calibration—Air Monitoring Instruments*. ANSI N323C-2009. New York: ANSI.

Ansoborlo, E., M. H. Henge-Napoli, V. Chazel, R. Gibert, and R. A. Guilmette. 1999. Review and critical analysis of available in vitro dissolution tests. *Health Phys.* 77(6):638–645.

Agency for Toxic Substances and Disease Registry (ATSDR). 1990. *Toxicological Profile for Radium*. Atlanta, GA: ATSDR.

Agency for Toxic Substances and Disease Registry (ATSDR). 1999. *Toxicological Profile for Uranium*. Atlanta, GA: ATSDR.

Agency for Toxic Substances and Disease Registry (ATSDR). 2007. *Toxicological Profile for Plutonium—Draft for Comment*. Atlanta, GA: ATSDR.

Agency for Toxic Substances and Disease Registry (ATSDR). 2008. *Toxicological Profile for Radon—Draft for Comment*. Atlanta, GA: ATSDR.

Baum, E. M., H. D. Knox, and T. R. Miller. 2003. *Nuclides and Isotopes: Chart of the Nuclides*, 16th Ed. Cincinnati: KAPL Inc. Lockheed Martin.

Biswas, P., and R. C. Flagan. 1988. The particle trap impactor. *J. Aerosol Sci.* 19:113–121.

Brunauer, S., P. H. Emmett, and E. Teller. 1938. Adsorption of gases in multimolecular layers. *J. Am. Chem. Soc.* 60(2):309–319.

Cember, H., and T. E. Johnson. 2009. *Introduction to Health Physics*, 4th Ed. New York: McGraw-Hill.

Chen, B. T., M. D. Hoover, G. J. Newton, S. J. Montano, and D. S. Gregory. 1999. Performance evaluation of the sampling head and annular kinetic impactor in the Savannah River Site alpha continuous air monitor. *Aerosol Sci. Technol.* 31:24–38.

Cheng, Y. S., R. A. Guilmette, Y. Zhou, J. Gao, T. LaBone, J. J. Whicker, and M. D. Hoover. 2004. Characterization of plutonium aerosol collected during an accident. *Health Phys.* 87(6):596–605.

Cohen, B. S. 2001. Radon and its short-lived decay product aerosols. In *Aerosol Measurement: Principles, Techniques, and Applications*, 2nd Ed. P. A. Baron and K. Willeke, eds. Hoboken, NJ: John Wiley and Sons, pp. 1011–1029.

Cohen, B. S., and M. L. Heikkinen. 2001. Sampling airborne radioactivity. In *Air Sampling Instruments for Evaluation of Atmospheric Contaminants*. 9th Ed. B. S. Cohen and C. S. McCammon, eds. Cincinnati: American Conference of Governmental Industrial Hygienists, pp. 221–240.

Cohen, B. S., M. Eisenbud, and N. H. Harley. 1980. Measurement of the α-radioactivity on the mucosal surface of the human bronchial tree. *Health Phys.* 39:619–632.

Dorrian, M. D., and M. R. Bailey. 1995. Particle size distributions of radioactive aerosols measured in workplaces. *Radiat. Protect. Dosim.* 60:119–133.

Eicholz, C. G., and J. S. Poston. 1979. *Principles of Nuclear Radiation Detection*. Ann Arbor, MI: Ann Arbor Science.

Eisenbud, M. 1987. *Environmental Radioactivity from Natural, Industrial, and Military Sources*. San Diego: Academic.

Evans, R. D. 1955. *The Atomic Nucleus*. New York: McGraw-Hill.

Evans, R. D. 1969. Engineers' guide to the elementary behavior of radon daughters. *Health Phys.* 17:229–252.

Finch, G. L., M. D. Hoover, J. A. Mewhinney, and A. F. Eidson. 1989. Respirable particle density measurements using isopycnic density gradient ultracentrifugation. *J. Aerosol Sci.* 20:29–36.

Firestone, R. B., ed. 1996. *Table of Isotopes*, 8th Ed. Berkeley, CA: Lawrence Berkeley National Laboratory (available online at http://ie.lbl.gov/toipdf/toi20.pdf).

Grivaud, L., S. Fauvel, and M. Chemtob. 1998. Measurement of performances of aerosol type radioactive contamination monitors. *Radiat. Prot. Dosim.* 79(1–4):495–498.

Hickey, E. E., G. A. Stoetzel, and P. C. Olsen. 1991. *Air Sampling in the Workplace*. NUREG 1400, Washington, DC: U.S. Nuclear Regulatory Commission.

Hobbs, C. H., and R. O. McClellan. 1986. Toxic effects of radiation and radioactive materials. In *Casarette and Doull's Toxicology: The Basic Science of Poisons*, 3rd Ed. C. D. Klaassen, M. O. Amdur, and J. Doull, eds. New York: MacMillan, pp. 669–705.

Hoover, M. D., B. T. Castorina, G. L. Finch, and S. J. Rothenberg. 1989. Determination of the oxide layer thickness on beryllium metal particles. *Am. Ind. Hyg. Assoc. J.* 50:550–553.

Hoover, M. D., A. F. Eidson, J. A. Mewhinney, G. L. Finch, B. J. Greenspan, and C. A. Cornell. 1988a. Generation and characterization of respirable beryllium oxide aerosols for toxicity studies. *Aerosol Sci. Tech.* 9:83–92.

Hoover, M. D., G. L. Finch, and B. T. Castorina. 1991. Sodium metatungstate as a medium for measuring particle density using isopycnic density gradient ultracentrifugation. *J. Aerosol Sci.* 22:215–221.

Hoover, M. D., C. J. Mewhinney, and G. J. Newton. 1999. Modular glovebox connector and associated good practices for control of radioactive and chemically toxic materials. *Health Phys.* 76:66–72.

Hoover, M. D., and G. J. Newton. 1991a. *Preliminary Evaluation of Optical Monitoring for Real-Time Detection of Radioactive Aerosol Releases*. Albuquerque, NM: Inhalation Toxicology Research Institute.

Hoover, M. D., and G. J. Newton. 1991b. Technical bases for selection and use of filter media in continuous air monitors for alpha-emitting radionuclides. In: *Annual Report of the Inhalation Toxicology Research Institute for 1990–1991, LMF-134*, Springfield, VA: National Technical Information Service, pp. 16–19.

Hoover, M. D., and G. J. Newton. 1992. Update on selection and use of filter media in continuous air monitors for alpha-emitting radionuclides. In: *Annual Report of the Inhalation Toxicology Research Institute for 1991–1992*, LMF-138. Springfield, VA: National Technical Information Service, pp. 5–7.

Hoover, M. D., and G. J. Newton. 1993. Radioactive aerosols. In *Aerosol Measurement: Principles, Techniques, and Applications*, 1st Ed. K. Willeke and P. A. Baron, eds. New York: Van Nostrand Reinhold.

Hoover, M. D., and G. J. Newton. 1998. Performance testing of continuous air monitors for alpha-emitting radionuclides. *Radiat. Prot. Dosim.* 79(1–4):499–504.

Hoover, M. D., G. J. Newton, R. A. Guilmette, R. J. Howard, R. N. Ortiz, J. M. Thomas, S. M. Trotter, and E. Ansoborlo. 1998. Characterisation of enriched uranium dioxide particles from a uranium handling facility. *Radiat. Prot. Dosim.* 79(1–4): 57–62.

Hoover, M. D., G. J. Newton, H. C. Yeh, and A. F. Eidson. 1983. Characterization of aerosols from industrial fabrication of mixed-oxide nuclear reactor fuels. In *Aerosols in the Mining and Industrial Work Environments*. V. A. Marple and B. Y. H. Liu, eds. Ann Arbor, MI: Ann Arbor Science.

Hoover, M. D., G. J. Newton, H. C. Yeh, F. A. Seiler, and B. B. Boecker. 1988b. *Evaluation of the Eberline Alpha-6 Continuous Air Monitor for Use in the Waste Isolation Pilot Plant: Phase I Report*. 21 December 1988. Albuquerque, NM: Inhalation Toxicology Research Institute.

Hoover, M. D., G. J. Newton, H. C. Yeh, F. A. Seiler, and B. B. Boecker. 1990. *Evaluation of the Eberline Alpha-6 Continuous Air Monitor for Use in the Waste Isolation Pilot Plant: Report for Phase II*. 31 January 1990. Albuquerque, NM: Inhalation Toxicology Research Institute.

International Commission on Radiological Protection (ICRP). 1975. *Reference Man: Anatomical, Physiological and Metabolic Characteristics*. International Commission on Radiological Protection Publication 23. Oxford: Pergamon Press.

International Commission on Radiological Protection (ICRP). 1979. *Limits on Intakes of Radionuclides by Workers*. International Commission on Radiological Protection Publication 30 and addendums. Oxford: Pergamon Press.

International Commission on Radiological Protection (ICRP). 1990. *1990 Recommendations of the International Commission on Radiation Protection*. International Commission on Radiological Protection Publication 60. *Ann. ICRP* 21(1/3). Oxford: Elsevier Science.

International Commission on Radiological Protection (ICRP). 1994a. *Human Respiratory Tract Model for Radiological Protection*. International Commission on Radiological Protection Publication 66. *Ann. ICRP* 24(4). Oxford: Elsevier Science.

International Commission on Radiological Protection (ICRP). 1994b. *Dose Coefficients for Intakes of Radionuclides by Workers*. International Commission on Radiological Protection Publication 68. *Ann. ICRP* 24(4). Oxford: Elsevier Science.

International Commission on Radiological Protection (ICRP). 2003. *Guide for the Practical Application of the ICRP Human Respiratory Tract Model*. International Commission on Radiological Protection Supporting Guidance 3. *Ann. ICRP* 32(1–2). Oxford: Elsevier Science.

International Electrotechnical Commission (IEC). 2002a. *Radiation Protection Instrumentation. Equipment for Continuously Monitoring Radioactivity in Gaseous Effluents, Part 1: General Requirements*, IEC Standard 61761-1. Geneva, Switzerland: IEC.

International Electrotechnical Commission (IEC). 2002b. *Radiation Protection Instrumentation. Equipment for Continuously Monitoring Radioactivity in Gaseous Effluents, Part 2: Specific Requirements for Radioactive Aerosol Monitors including Transuranic Aerosols*, IEC Standard 61761-2. Geneva, Switzerland: IEC.

International Electrotechnical Commission (IEC). 2002c. *Radiation Protection Instrumentation. Equipment for Continuously Monitoring Radioactivity in Gaseous Effluents, Part 3: Specific Requirements for Radioactive Noble Gas Monitors*, IEC Standard 61761-3. Geneva, Switzerland: IEC.

International Electrotechnical Commission (IEC). 2002d. *Radiation Protection Instrumentation. Equipment for Continuously Monitoring Radioactivity in Gaseous Effluents, Part 4: Specific Requirements for Radioactive Iodine Monitors*, IEC Standard 61761-4. Geneva, Switzerland: IEC.

International Electrotechnical Commission (IEC). 2002e. *Radiation Protection Instrumentation. Equipment for Continuously Monitoring Radioactivity in Gaseous Effluents, Part 5: Specific Requirements for Tritium Effluent Monitors*, IEC Standard 61761-5. Geneva, Switzerland: IEC.

International Electrotechnical Commission (IEC). 2007. *Radiation Protection Instrumentation. Equipment for Monitoring Radioactive Noble Gases*, IEC Standard 62302. Geneva, Switzerland: IEC.

International Electrotechnical Commission (IEC). 2008. *Radiation Protection Instrumentation. Equipment for Monitoring Airborne Tritium*, IEC Standard 62303. Geneva, Switzerland: IEC.

Kanapilly, G. M., O. G. Raabe, C. H. T. Goh, and R. A. Chimenti. 1973. Measurement of the *in vitro* dissolution of aerosol particles for comparison to *in vivo* dissolution in the respiratory tract after inhalation. *Health Phys.* 24:497–507.

Knoll, G. F. 2000. *Radiation Detection and Measurement*. New York: John Wiley and Sons.

Kotrappa, P. and M. E. Light. 1972. Design and performance of the Lovelace aerosol particle separator. *Rev. Sci. Instrum.* 43:1106–1112.

L'Annunziata, M. F., ed. 2003. *Handbook of Radioactivity Analysis*, 2nd Ed. London: Academic.

Lucas, H. F. 1977. Alpha scintillation counting. in *Atomic Industrial Forum Workshop on Methods for Measuring Radiation in and around Uranium Mills*, Vol. 3, No. 9, E. D. Harwood, ed., Washington, DC: Atomic Industrial Forum.

Lindekin, C. L., F. K. Petrock, W. A. Phillips, and R. D. Taylor. 1964. Surface collection efficiency of large-pore membrane filters. *Health Phys.* 10:495–499.

MacArthur, D. W., and K. S. Allander. 1991. *Long-Range Alpha Detectors*. LA-12073-MS. Los Alamos, NM: Los Alamos National Laboratory.

Maiello, M. L., and M. D. Hoover, eds. 2011. *Radioactive Air Sampling Methods*. Boca Raton, FL: CRC Press.

Martell, E. A. 1975. Tobacco radioactivity and cancer in smokers. *Am. Sci.* 63: 404–412.

McFarland, A. R. 1998. *Methodology for Sampling Effluent Air from Stacks and Ducts of the Nuclear Industry*. LA-UR-96-2958, Revised July 1998, Los Alamos, NM: Los Alamos National Laboratory.

McFarland, A. R., C. A. Ortiz, M. E. Moore, R. E. DeOtte, Jr., and A. Somasundaram. 1989. A shrouded aerosol sampling probe. *Environ. Sci. Technol.* 23:1487–1492.

McFarland, A. R., J. C. Rodgers, C. A. Ortiz, M. E. Moore. 1992. A continuous sampler with background suppression for monitoring alpha-emitting aerosol particles. *Health Phys.* 62: 400–406.

McFarland, A. R., N. K. Anand, C. A. Ortiz, R. Gupta, S. Chandra, and A. P. McManigle. 1999a. A generic mixing system for achieving conditions suitable for single point representative effluent air sampling. *Health Phys.* 76:17–26.

McFarland, A. R., R. Gupta, and N. K. Anand. 1999b. Suitability of air sampling locations downstream of bends and static mixing elements. *Health Phys.* 77:703–712.

Mercer, T. T. 1967. On the role of particle size in the dissolution of lung burdens. *Health Phys.* 13:1211–1223.

Mercer, T. T. 1973. *Aerosol Technology in Hazard Evaluation*. New York: Academic.

Mercer, T. T., M. I. Tillery, and G. J. Newton. 1970. A multi-stage low flow rate cascade impactor. *J. Aerosol Sci.* 1: 9–15.

Mewhinney, J. A. 1978. *Radiation Exposure and Risk Estimates for Inhaled Airborne Radioactive Pollutants including Hot Particles, Annual Progress Report, July 1, 1976–June 30, 1977*. NUREG/CR-0010, Albuquerque, NM: Inhalation Toxicology Research Institute.

Mewhinney, J. A., A. F. Eidson, and V. A. Wong. 1987a. Effect of wet and dry cycles on dissolution of relatively insoluble particles containing Pu. *Health Phys.* 53:337–384.

Mewhinney, J. A., S. J. Rothenberg, A. F. Eidson, G. J. Newton, and R. Scripsick. 1987b. Specific surface area determination of U and Pu particles. *J. Colloid Interf. Sci.* 116:555–562.

Mine Safety and Health Administration (MSHA). 1971. Title 30, Code of Federal Regulations, Part 57. *Safety and Health Standards Underground Metal and Nonmetal Mines*. Washington, DC: U.S. Department of Labor. Mine Safety and Health Administration.

Moore, M. E., A. R. McFarland, and J. C. Rodgers. 1993. Factors that affect alpha particle detection in continuous air monitor applications. *Health Phys.* 65:69–81.

Morrow, P. E. and T. T. Mercer. 1964. A point-to-plane electrostatic precipitator for particle size sampling. *Am. Ind. Hyg. Assoc. J.* 25:8–14.

National Academy of Sciences (NAS). 1999. *Health Effects of Exposure to Radon: BEIR VI*. Washington, DC: National Academy Press.

National Council on Radiation Protection and Measurements (NCRP). 1978a. *Instrumentation and Monitoring Methods for Radiation Protection*. NCRP Report No. 57. Bethesda, MD: NCRP.

National Council on Radiation Protection and Measurements (NCRP). 1978b. *A Handbook of Radiation Protection Measurements Procedures*. NCRP Report No. 58. Bethesda, MD: NCRP.

National Council on Radiation Protection and Measurements (NCRP). 1988. *Measurement of Radon and Radon Daughters in Air*. NCRP Report No. 97. Bethesda, MD: NCRP.

National Council on Radiation Protection and Measurements (NCRP). 1997. *Deposition, Retention and Dosimetry of Inhaled Radioactive Substances*. NCRP Report No. 125. Bethesda, MD: NCRP.

National Council on Radiation Protection and Measurements (NCRP). 2009. *Ionizing Radiation Exposure of the Population of the United States*, NCRP Report No. 160. Bethesda, MD: NCRP.

National Institute for Occupational Safety and Health (NIOSH). 1987. *A Recommended Standard for Occupational Exposure to Radon Progeny in Underground Mines*. Washington, DC: National Institute for Occupational Safety and Health, DHHS (NIOSH) Publication No. 88–101.

Newton, G. J., M. D. Hoover, E. B. Barr, B. A. Wong, and P. D. Ritter. 1987. Collection and characterization of aerosols from metal cutting techniques typically used in decommissioning nuclear facilities. *Am. Ind. Hyg. Assoc. J.* 48:922–932.

Papastefanou, C. 2007. *Radioactive Aerosols*. Amsterdam: Elsevier.

Parkhurst, M. A., E. G. Daxon, G. M. Lodde, F. Szrom, R. A. Guilmette, L. E. Roszell, G. A. Fallo, and C. B. McKee. 2005. *Depleted Uranium Aerosol Doses and Risks: Summary of U.S. Assessments*. Richland, WA: Battelle Press.

Parkhurst, M. A., R. A. Guilmette, T. D. Holmes, Y. S. Cheng, M. D. Hoover, F. Szrom, G. A. Falo, J. J. Whicker, D. P. Alberth, J. L. Kenoyer, R. J. Traub, K. M. Krupka, K. Gold, B. W. Arey, E. D. Jenson, G. Miller, T. T. Little, L. E. Roszell, F. F. Hahn, B. Robyn, E. G. Daxon, G. M. Lodde, and M. A. Melanson. 2009. Special issue on the capstone depleted uranium aerosol characterization and human health assessment. *Health Phys.* 96(3):207–409.

Price, W. S. 1965. *Nuclear Radiation Detection*. New York: McGraw-Hill.

Raabe, O. G. 1972. Instruments and methods for characterizing radioactive aerosols. *IEEE Trans. Nucl. Sci.* NS-19(1):64–75.

Raabe, O. G., G. J. Newton, C. J. Wilkenson, and S. V. Teague. 1978. Plutonium aerosol characterization inside safety enclosures at a demonstration mixed-oxide fuel fabrication facility. *Health Phys.* 35:649–661.

Riehl, J. R., V. R. Dileep, N. K. Anand, and A. R. McFarland. 1996. *DEPOSITION 4.0: An Illustrated User's Guide*. Aerosol Technology Laboratory Report 8838/7/96. Department of Mechanical Engineering. College Station, Texas: Texas A&M University.

Rodgers, J. C., C. I. Fairchild, G. O. Wood, C. A. Ortiz, A. Muyshondt, and A. R. McFarland. 1996. Single point aerosol sampling: Evaluation of mixing and probe performance in a nuclear stack. *Health Phys.* 70:25–35.

Rothenberg, S. J., P. B. Denee, Y. S. Cheng, R. L. Hanson, H. C. Yeh, and A. F. Eidson. 1982. Methods for the measurement of surface areas of aerosols by adsorption. *Adv. Colloid Interf. Sci.* 15:223–249.

Rothenberg, S. J., D. K. Flynn, A. F. Eidson, J. A. Mewhinney, and G. J. Newton. 1987. Determination of specific surface area by krypton adsorption, comparison of three different methods of determining surface area, and evaluation of different specific surface area standards. *J. Colloid Interf. Sci.* 116:541–554.

Ruzer, L. S. 2005. Radioactive aerosols. In *Aerosols Handbook−Measurement, Dosimetry, and Health Effects*. L. S. Ruzer and N. H. Harley, eds. Boca Raton: CRC.

Schayer, S., B. Nowak, Y. Wang, Q. Qu, and B. Cohen. 2009. ^{210}Po and ^{210}Pb activity in Chinese cigarettes. *Health Phys.* 96(5):543–549.

Schery, S. D. 2001. *Understanding Radioactive Aerosols and Their Measurement*. Boston: Kluwer.

Schleien, B. S., L. A. Slabeck, Jr., and B. K. Kent, eds. 1998. *Handbook of Health Physics and Radiological Health*, 3rd Ed. Baltimore, MD: Lippincott Williams & Wilkins.

Scott, B. R., and A. F. Fencl. 1999. Variability in PuO$_2$ intake by inhalation: Implications for worker protection at the US Department of Energy. *Radiat. Protect. Dosim.* 83:221–232.

Scott, B. R., M. D. Hoover, and G. J. Newton. 1997. On evaluating respiratory tract intake of high-specific activity emitting particles for brief occupational exposure. *Radiat. Protect. Dosim.* 69(1):43–50.

Shapiro, J. 1981. *Radiation Protection, a Guide for Scientists and Physicians*. Cambridge: Harvard University Press.

Stabin, M. G. 2008. *Radiation Protection and Dosimetry: An Introduction to Health Physics*. New York: Springer Science and Business Media.

Stefaniak, A. B., R. A. Guilmette, G. A. Day, M. D. Hoover, P. N. Breysse, and R. C. Scripsick. 2005. Characterization of phagosomal simulant fluid for study of beryllium aerosol dissolution. *Toxicol. In Vitro* 19(1):123–134.

Stefaniak, A. B., C. A. Brink, R. M. Dickerson, G. A. Day, M. J. Brisson, M. D. Hoover, and R. C. Scripsick. 2007. A theoretical framework for evaluating analytical digestion methods for poorly soluble particulate beryllium. *Anal. Bioanal. Chem.* 87(7): 2411–2417.

Stöber, W., and H. Flachsbart. 1969. Size-separating precipitation of aerosols in a spinning spiral duct. *Environ. Sci. Techol.* 3:1280–1296.

Tait, G. W. C. 1956. Determining concentration of airborne plutonium dust. *Nucleonics* 14:53–55.

Teague, S. V., H. C. Yeh, and G. J. Newton. 1978. Fabrication and use of krypton-85 aerosol discharge devices. *Health Phys.* 35:392–395.

Turner, J. E. 1996. *Atoms, Radiation, and Radiation Protection*, 2nd Ed. New York: John Wiley & Sons.

Turner, J. E., J. S. Bogard, J. B. Hunt, and T. A. Rhea. 1988. *Problems and Solutions in Radiation Protection*. New York: Pergamon.

Unruh, W. P. 1986. *Development of a Prototype Plutonium CAM at Los Alamos*. LA-UR-90-2281. December 15, 1986. Los Alamos, NM: Los Alamos National Laboratory.

U.S. Department of Energy (USDOE). 1988. *Operational Health Physics Training*. ANL-88-26. Prepared by Argonne National Laboratory, Argonne, IL, for the U.S. Department of Energy Assistant Secretary for Environment, Safety, and Health. Oak Ridge, TN: National Technical Information Service.

U.S. Department of Energy (USDOE). 1994. *Airborne Release Fractions/Rates and Respirable Fractions for Nonreactor Nuclear Facilities*, DOE-HDBK-3010-94, vols. 1 and 2. December 1994, Washington, DC: U.S. Department of Energy.

U.S. Department of Energy (USDOE). 1998. *The Department of Energy Laboratory Accreditation Program for Radiobioassay*, DOE STD 1112-98. December 1998, Washington, DC: U.S. Department of Energy.

U.S. Department of Energy (USDOE). 2007. *Occupational Radiation Protection*. Title 10, Code of Federal Regulations, Part 835. June 8, 2007. Washington, DC: U.S. Government Printing Office.

U.S. Environmental Protection Agency (USEPA). 2003a. *EPA Assessment of Risks from Radon in Homes*. EPA-402-R-03-003. June 2003. Washington, DC: U.S. Government Printing Office.

U.S. Environmental Protection Agency (USEPA). 2003b. *National Emission Standards for Hazardous Air Pollutants*. Title 40, Code of Federal Regulations, Part 61. July 1, 2003. Washington, DC: U.S. Government Printing Office.

U.S. Environmental Protection Agency (USEPA). 2006. *Inventory of Radiological Methodologies for Sites Contaminated with Radioactive Materials*, EPA-402-R-06-007. October 2006. Washington, DC: U.S. Government Printing Office.

U.S. Nuclear Regulatory Commission (USNRC). 2006. *Standards for Protection Against Radiation*. Title 10, Code of Federal Regulations, Part 20 et al. January 1, 2006. Washington, DC: U.S. Government Printing Office.

U.S. Nuclear Regulatory Commission (USNRC). 1992. *Calibration and Error Limits of Air Sampling Instruments for Total Volume of Air Sampled*. Regulatory Guide 8.25. Washington, DC: U.S. Nuclear Regulatory Commission.

U.S. Nuclear Regulatory Commission (USNRC). 2000. *The Regulation and Use of Radioisotopes in Today's World*. NUREG/BR-0217. Washington, DC: U.S. Nuclear Regulatory Commission.

Voigts, Chr., G. Siegmon, M. Berndt, and W. Enge. 1986. Single alpha-emitting aerosol particles. In *Aerosols, Formation and Reactivity*, 2nd International Aerosol Conference, Berlin. Oxford: Pergamon, p. 1153.

Yeh, H. C., G. J. Newton, O. G. Raabe, and D. R. Boor. 1976. Self-charging of ^{198}Au-labeled monodisperse gold aerosols studied with a miniature electrical mobility spectrometer. *J. Aerosol Sci.* 7:245–253.

Yeh, H. C., G. J. Newton, and S. V. Teague. 1978. Charge distribution on plutonium-containing aerosols produced in mixed-oxide reactor fuel fabrication and the laboratory. *Health Phys.* 35:500–503.