

# Sampling For Airborne Radioactivity\*

J. M. HESLEP AND A. W. BELLAMY  
Radiological Safety Services  
State of California, Office of Civil Defense  
Sacramento, California

As the nation's industrial complex grows, one of its attendant evils—environmental pollution—grows apace. Perhaps no segment of this complex grows more rapidly at present than the atomic energy industry. Responsible officials are gradually becoming aware of the potential for pollution of this new giant. The problem is complicated by security requirements and by virtue of the inability to sense the presence of radioactive contaminants. The present contribution considers some of the general aspects of one segment of this relatively new problem—airborne radioactive contamination.

## Nature of Radioactivity

Atoms which are unstable as a result of the relative numbers or configurations of the constituents of their nuclei are said to be radioactive. In the process of achieving greater stability, these nuclei undergo a process of disintegration, one of the results of which is the emission of energy in the form of radiation. Based on its origin, this is termed nuclear radiation, to distinguish from other types of radiations which result from extra-nuclear phenomena. It is important to distinguish between the source of the nuclear radiation (the radioactive material) and the radiation emitted therefrom.

Most chemical elements have several forms, differing in the mass of the atomic nuclei, but not in chemical properties. These different forms of the same element are called isotopes. There are over 300 naturally occurring isotopes of some 90 chemical elements. Of these, about 16% are radioactive, the remainder stable. In addition, there is a growing list of over 600 different radioactive isotopes which have been produced artificially—all in the last 15 years.

Each radioactive isotope has distinctive properties in the way of type and energy of radiation emitted, and rate of decay. The latter property is commonly characterized in terms of half-life—the time required for one half of a given number of atoms to undergo disintegration. Half-lives of the various isotopes vary from fractions of a second to billions of years.

There are several types of nuclear radiations. We need consider three types—alpha, beta, and gamma. Alpha and beta radiations are composed of fast-moving, discrete particles—helium nuclei and electrons, respectively. Gamma radiation is nonparticulate, being electromagnetic

in character, and differing from visible light only in wave length, and therefore energy.

The primary effect of the various types of radiation is the same—the transfer of energy to the medium traversed, usually resulting in ionization, or excitation. For this reason, nuclear radiations are often referred to also as ionizing radiations. Nuclear radiations are not detectable by any of the human senses. Detection and measurement are made possible by use of instruments which measure some effect of the radiation—commonly, ionization. Ionization is also the principle basis for damage to biological systems by nuclear radiations.

Due to differences in range of travel and specific ionization, the various types of radiation vary in their biological effectiveness. Alpha particles are relatively heavy and highly charged. Thus, their penetration is very low, but their specific ionization (ionization produced per unit path length) is very high. Beta particles are much lighter and lesser charged. Their penetration is moderate. Gamma rays are weightless, chargeless, and highly penetrating. The amount of energy associated with emitted radiation varies between isotopes. Obviously, the greater the energy of the radiation of a given type, the greater its biological effectiveness. These properties have an important bearing on both detection and biological effectiveness.

## Biological Effectiveness of Nuclear Radiations

This is not the place to consider basic mechanisms of radiation damage to biological systems. It can be generalized that the biological effect is directly proportional to the radiation dose per unit volume and to the radiosensitivity of the body organ irradiated.

There exist 2 rather different categories of hazard. The first of these, which we shall only mention, occurs when the source of radiation remains outside the body. This is referred to as the external radiation hazard. Gamma radiation is the most important type here, due to its high penetrability. Beta radiation affects the surface layers of the body (skin) only, although this can be serious. Alpha radiation is generally considered to afford no external hazard. The distinctive feature is the ability to eliminate the external hazard by removing the source of radiation from the proximity of the person, or vice versa.

The other category of hazard is internal—i.e., when the source of radiation is taken into the body. In this case it is more difficult to separate source and object. Individual radioactive isotopes vary in their degree of internal

\*Presented at the First Semi-Annual Technical Conference of the Air Pollution Control Association at Los Angeles, California, November 4-6, 1954.

hazard, depending on: (1) type of radiation emitted, (2) energy of the radiation, (3) rate of decay of the material, (4) distribution of the material in the body, and (5) the rate and completeness of excretion of material from the body. The latter two factors are functions of the chemical nature and the physical state of the contaminating material.

The relative internal hazard of the 3 types of radiation is just the reverse of the external case. Intense damage is effected in a small volume around sources of alpha radiation. Where these sources reside in critical body organs, an extreme hazard may result. Beta radiations are again intermediate, but of generally greater concern than from external sources. Gamma radiation is of lesser significance, except for the difficulty of removing the source.

It is perhaps worthwhile to interject at this point the fact that the body is continuously being subjected to nuclear radiations from cosmic rays and naturally occurring radioactive materials. Radiation exposure is nothing new. The rapid growth of the atomic energy industry, however, adds greatly to the potential for exposure.

The 2 principle routes of radioactive materials into the body are, of course, ingestion and inhalation. There is reason to believe that, except under most unusual conditions, inhalation offers the more significant potential.<sup>1</sup> The remaining discussion will be devoted to the significance of radioactive materials as air pollutants, and specifically as this relates to an inhalation hazard.

The distinguishing characteristic of a radioactive aerosol is its extreme toxicity. Maximum permissible concentrations represent fantastically small amounts of matter, being of the order of  $\mu\text{-}\mu\text{g}/\text{m}^3$  for some isotopes. This compares with measurement of many chemo-toxic materials in  $\text{mg}/\text{m}^3$ . Of course, radioactive particulates are mixed environmentally with much larger amounts of non-active materials.

Aside from the nature of the particular radioisotope which is inhaled, the size and solubility of the particles of which it is a part are important. Radioactive particles behave in the same way as other particles in being deposited in the respiratory system. In the nasal system, all particles larger than about  $5\ \mu$  diameter are filtered out. The efficiency of nasal retention decreases with particle size, reaching essentially zero for  $1\ \mu$  particles and smaller. Over-all respiratory retention is virtually complete for about  $5\ \mu$  particles, and decreases to a minimum of some 25% for  $0.25\ \mu$  particles. The lower retention of fine particles is attributed at least partially to the fact that they may be inhaled, then exhaled again without ever impinging on any surface.<sup>2</sup> Currently accepted standards for maximum permissible concentrations of air-

borne radioactivity<sup>3</sup> are based on the following assumptions regarding retention of inhaled particulates: (1) In the case of soluble compounds, 25% is retained in the lower respiratory tract. From this point it goes to the blood stream, its fate within the body then depending on the chemical nature of the material. Further, another 50% is held in the upper respiratory tract and subsequently swallowed. (2) In the case of insoluble compounds, it is assumed that 12% is retained in the lower respiratory tract. The rest is exhaled or swallowed.

In performing necessary computations, one must make assumptions, and the ones made are probably as good as any for general use. There is evidence, however, that particle size has a marked effect on body retention. One study<sup>4</sup>, for example, indicates that retention of insoluble particles may vary from 1.2 to 38%, depending on the particle size distribution.

Further evidence of the importance of the particle size factor is shown by the following statement<sup>5</sup>: "A maximum permissible air concentration for mixed fission products following a nuclear detonation has been established by the U. S. AEC upon the recommendation of an advisory panel of experts. This concentration is 100 micromicrocuries/ $\text{m}^3$  of air, averaged over a 24-hr. period. Since particles between  $\frac{1}{2}$  and  $5\ \mu$  are most likely to be retained in the lung, the maximum permissible concentration is 1 micromicrocurie/ $\text{m}^3$  of air if the radioactivity is associated with particles less than  $5\ \mu$  in diameter."

Once inside the body, the site of deposition of the radioactive material assumes some importance. Frequently, these materials tend to concentrate in certain specific organs of the body. For that reason, calculations of resulting exposure are made on the basis of the body organ receiving the greatest radiation dose, the so-called "critical organ." For inhaled insoluble particulates, the lung itself is the critical organ. For soluble material, the chemical nature determines the critical organ. The most striking single example is concentration of iodine in the thyroid. As an aside, it might be pointed out that widespread use is made of this phenomenon in medical diagnosis, therapy and research.

#### Exposure to Airborne Radioactive Materials

We might now examine the potential sources of airborne radioactive particulates, and indicate who might be exposed to them. Radioactive aerosols are produced in the atomic energy industry: (1) as dusts in the cooling air from some nuclear reactors; (2) as mists and fumes arising during the separation of plutonium from fission products; (3) as dusts from the mining, transport-

(1) Eisenbud, M., Blatz, H., and Barry, E. V., "How Important Is Surface Contamination," *Nucleonics*, 12, No. 8, 12-15 (1954).

(2) Morgan, G. W., and Buchanan, C. R., "Air Contamination and Respiratory Protection in Radioisotope Work," *AECU Report 2821* (1953).

(3) "Maximum Permissible Amounts of Radioisotopes in the Human Body and Maximum Permissible Concentrations in Air and Water," National Bureau of Standards Handbook No. 52 (1953).

(4) Burnett, T. J., "Sampling Methods and Requirements for Estimating Airborne Radioparticulate Hazards," *ORNL Report CF-52-11-1* (1952).

(5) "Assuring Public Safety in Continental Weapons Tests," *Thirteenth Semi-Annual Report of the U.S. Atomic Energy Commission* (1953).

ing, and storing of uranium ore; (4) as dusts and mists in manipulating and processing radioactive ores; (5) during incineration of radioactive wastes and by-products; (6) during laboratory and plant operations in various phases of atomic energy work; and (7) during detonation of atomic weapons.

Potential exposure from these sources may be considered in 3 categories. First, there are those persons working in mines, plants, or laboratories where radioactive aerosols are produced. In the second category are the areas immediately adjacent to such installations, for distances up to several miles. Lastly, we might consider areas extending over hundreds or thousands of square miles.

We may state categorically that the Atomic Energy Commission and its contractors have done an outstanding job in protecting the people who work in their installations and those who live nearby. The safety record has been little short of phenomenal in view of what might have happened.

Widespread environmental contamination is a more complex problem. Insofar as we have knowledge, the only appreciable source of widespread contamination is atomic weapons testing. We do not, as yet, know with certainty what additional problems of this type will confront us with the vast expansion of the atomic energy industry which will undoubtedly occur in the foreseeable future. We certainly do not know all of the things that will happen in the event that atomic weapons are used in war on our country. There is a strong suspicion that inhalation and ingestion of radioactive materials may represent a serious, long-range problem.

#### Sampling and Measurement of Airborne Radio-particulates

To this point, there has been an attempt to point out some of the reasons why airborne radioactivity is of concern. It is obvious that if hazards from this source are to be recognized and controlled, there must be some mechanism for collection of adequate samples and for making appropriate determinations of the content thereof.

Sampling is a problem, the difficultness of which is shared by all phases of scientific endeavor. The universal, and never quite attainable, goal is that phantom — the "representative sample." Even the term is difficult of definition. In its simplest sense, it means to obtain an aliquot which is identical, in all of its important (for the purpose at hand) aspects, with the population which is of interest to the sampler. Every sample is representative of something. The essential question is—how far into the medium as a whole is it safe to extrapolate results obtained on a given sample of the medium? Statistics is a useful tool as an assistant; still, much is left to be desired.

Sampling of the air for particulate contamination represents one of the more difficult of sampling problems. Efficient samplers have been developed, but the problem of extrapolation is often acute. For example, in a laboratory or plant there frequently exist vast differences in

concentrations of pollutants over limited distances. Environmental sampling is made complex because of the vagaries of micro-meteorology. In spite of this, much useful information is obtainable. Inherent limitations, however, must never be overlooked.

The simplest and most widely used type of sampler for airborne particulates is one based on concentration of the sample by filtering. Any device which can be used to pull air through an appropriate filtering material is satisfactory. In the authors' laboratory, relatively inexpensive commercial vacuum cleaners and Hollingsworth and Vose H-70 filter paper are used, with rather good results.

The critical features of this type of sampler are: (1) the efficiency of the filtering material, (2) particle size of contaminants, and (3) the atmospheric concentration of contaminants. It is quite common to obtain sampling efficiencies of 95 to 99% with filter paper samplers. Use of the so-called molecular, or membrane, filters now available should increase this efficiency, although not representing the perfect solution.

Other types of samplers in common use are electrostatic precipitators, impingers, and cyclone separators. Each has its advantages for specific problems, and disadvantages for universal use.

All of the samplers so far mentioned are more or less useful in determining gross concentrations of contaminants over a fairly wide range of particle size. The importance of the particle size factor has been pointed out. It is, thus, frequently desirable to ascertain the particle size distribution of airborne contaminants. Again, no single method or technique can provide adequate information of this type in all circumstances.

Optical and electron microscopes are used, but procedures are relatively tedious, and techniques not easily adapted for routine work. The thermal precipitator is quite useful under certain conditions in determination of very small particles.

Perhaps the most widely useful sampler for particle size determinations is the cascade impactor. This instrument can be considered as a sort of multiple jet sampler. It employs a series of jets of decreasing areas and collecting slides arranged in tandem. The first stage removes the largest particles, and each succeeding stage progressively removes a segment of the upper size range of the remaining distribution. The usual model has 5 stages, the final one of which is a filter paper. Good results are obtained in the range between 0.2 and 50  $\mu$  diameter. Below 0.3 to 0.6  $\mu$  sampling is efficient but there is not good characterization of particle size.<sup>6</sup> There are difficulties of calibration and so on, but by and large this is a good instrument for the purpose.

Once a sample has been obtained, the problem becomes one of analysis for radioactive content and of evaluation

(<sup>6</sup>) Stokinger, H. E., and Laskin, S., "Air Pollution and the Particle Size Toxicity Problem," *Nucleonics*, 6, No. 3, 15-31 (1950)

of possible hazards. In addition to such parameters as volume sampled, representativeness of sample, and particle size distribution, a determination of the isotopic species present must be made. It is also desirable to determine the physical and chemical nature of the particulate material, but only when the exact source of the pollutant is known can this be evaluated without elaborate analyses.

If the collected sample is sufficiently active (in radioactive content) and provided a complex mixture of isotopes is not present, identification of specific contaminants is usually possible. This is done by measurement of type and energy of radiation emitted and of the rate of decay (half-life). One must then equate the results with accepted values for maximum permissible concentrations of each isotope present.

One additional complication is the presence in the atmosphere of naturally occurring radioactive materials. These are principally the radioactive gases (radon and thoron) and their decay products. They become adsorbed on dust particles and are collected by the air sampler. The activity from this source can cause considerable complication in interpretation of data on the content of artificial contaminants if the determinations are made soon after sample collection. By allowing the sample to stand for about 24 hr. between collection and measurement, however, this complication is effectively eliminated. Most artificial contaminants of interest have half-lives of sufficient length to allow this procedure to be followed.

If the atmospheric concentration of these natural contaminants were relatively constant, appropriate corrections could be made for them. Such is not the case; and the fact that their concentration varies between rather wide limits is of considerable interest in certain meteorological investigations.

Some airborne radioactivity samplers are in use in which the detection and measurement equipment is made an integral part of the apparatus. Filter paper is inserted in a roll. Air is drawn through a segment of the paper. After a given interval of time, the paper is automatically caused to move, so that the exposed segment is placed near an appropriate detecting element, and a new segment of paper is exposed. A recording mechanism may be attached to the detector, making an automatic, continuously operating assembly.

Still another technique involves passing air directly into a measuring chamber, without concentration.

#### Constructive Use of Radioactive Aerosols

Aside from an interest in the possible hazards of airborne radioactivity, a potential exists for making constructive use of the phenomena involved, particularly in meteorological research. It is feasible to "tag" air currents with radioactive aerosols, and to study the subsequent behavior of these currents by tracking the radioactivity. This should be possible with amounts of material well below hazardous

concentrations. Meteorological information could be obtained with this technique that could probably be gotten in no other way. Some caution is indicated, however, primarily because any additional sources of environmental radioactive contamination must be carefully scrutinized, and a balance struck between advantages and disadvantages.

Some of the possibilities are indicated by work of the authors' laboratory. These indications are on a very gross basis, as no control could be exercised over the important factors involved.

During the last 3 atomic weapons testing programs in Nevada, we have operated a statewide air-sampling program. From 10 to 20 rather widely scattered sampling stations have been used. After the announcement of each test shot, a prediction was made of the general path that the radioactive residue should follow. Predictions were based on meteorological data available from regular U. S. Weather Bureau sources. Observations from the air samplers were then compared with the meteorological predictions. There have been handicaps in that we could sample only in California, and prevailing winds are such that most of the material goes to the east and northeast, away from California.

Nevertheless, some interesting results were obtained. Of 25 test shots of which there is definite knowledge, radioactive material from about 16 was detected at 1 or more of our sampling stations. The amount and distribution of material has been highly variable. A qualitative correlation was made between prediction and observation for each test. The correlation was classified as good to excellent in 18 cases and as fair to poor in 7. In general, as may be expected, predictions were reasonably accurate when wind velocities were moderate to high, and when winds at all elevations were from the same general direction. In other cases, results differed appreciably from predictions. There were exceptions, however, to both examples.

#### Conclusions

The rapid growth of the atomic energy industry adds another dimension to the field of environmental pollution, but there is, at present, no evidence to indicate cause for acute widespread concern over the problem. It is not too early, however, to develop plans to cope with an acute situation, should it arise. As a minimum, widespread periodic sampling for airborne radioactivity is indicated.

As the base of the atomic energy industry broadens, and as operating costs become more of a factor, constant vigilance must be maintained to assure that the same high safety standards now prevalent are maintained.

Nuclear energy offers much that can serve to better man's lot. However, it is incumbent upon responsible officials and agencies to insure that the populace is protected from associated hazards, interfering as little as possible with development of beneficial aspects.