Samples taken in 1955 indicate that most atmospheric particles likely to reach the lungs had a radioactivity of less than 5 μμc., but the radioactivity of single particles was as high as 500 μμc. The average concentration of beta activity was about 5 μμc. per cubic meter of air.

Radioactive Particles in the Atmosphere at Cincinnati, Ohio

R. LOUIS BRADSHAW, M.S.E.E., and LLOYD R. SETTER, Ph.D.

THE RADIOACTIVITY of air particles in samples collected in 20 to 30 municipalities in the National Air Sampling Network, established in 1953, indicated a thousandfold or more increase in beta radioactivity immediately following known tests of nuclear weapons. It was suspected that substantial amounts of the total radioactivity might be associated with discrete particles which might find their way into the respiratory tract of man. To determine the distribution of radioactivity in atmospheric particulate matter, a study was made of air samples collected at the Robert A. Taft Sanitary Engineering Center, Cincinnati, Ohio, during the winter and spring of 1955.

Sampling Apparatus

The particles in air samples were separated into primary and secondary fractions by a cyclone separator (*A*) in series with a filter (fig. 1). An air sampling rate of approximately 40 cubic feet per minute was selected, since at this flow the unit has an efficiency versus particle-size curve comparable to that of the upper respiratory system in man (*1*, *2*).

A nosepiece on the intake of the cyclone separator provided a face velocity of 25 liters per minute per square centimeter, which approximates the nasal air velocity of a man at work. Particles passing through the separator were caught on the glass fiber filter. These, constituting the secondary fraction, are the particles that might penetrate to the alveoli in actual breathing. They were deposited on an effective filter area of 63 square inches.

Procedure

The sampler was placed on the roof terrace off the east side of the auditorium of the Sanitary Engineering Center, approximately three stories above the ground. The sampling rate was adjusted by a bleeder valve on the high-volume blower (capacity of 60–80 cubic feet per minute) to 40.5 cubic feet per minute at the beginning of each sampling period. The sampling rate at 24 hours was rarely more than 10 percent below the initial rate. The average of the initial and final rates was taken for flow calculation.

Two samples were collected February 16–18, 1955, before the first United States test of nuclear weapons that month. From February

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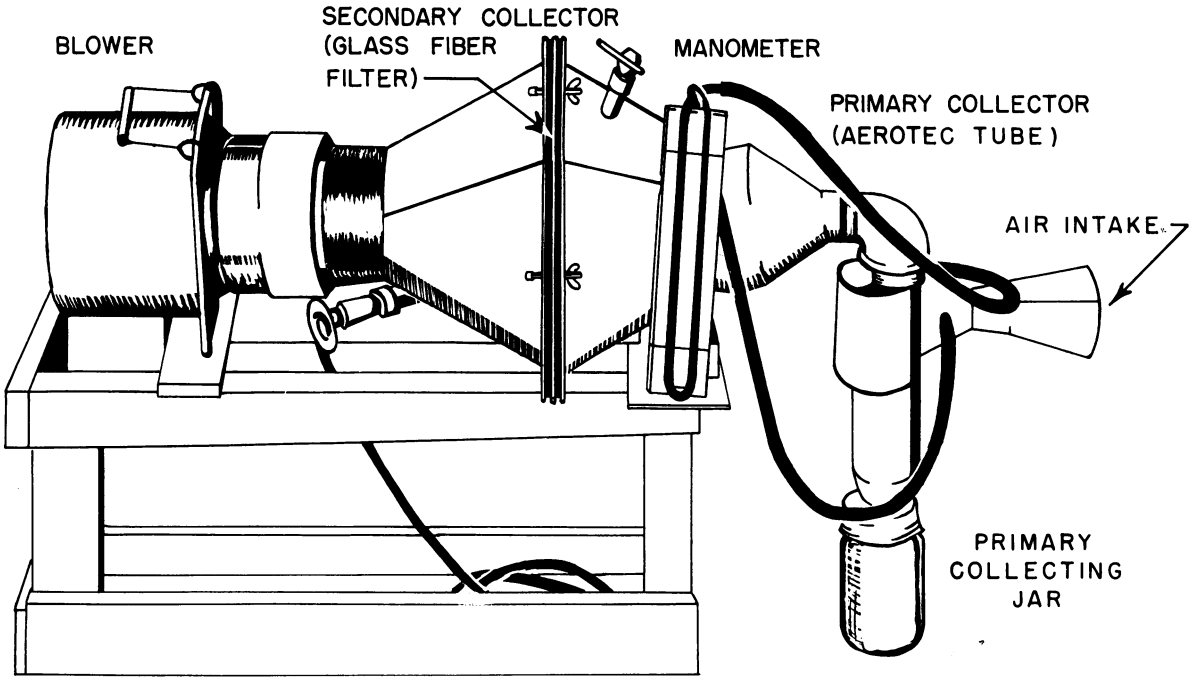


Figure 1. Air sampler used in Cincinnati study of atmospheric radioactivity, 1955.

19 through April 9 and again from May 5 through May 12, sampling was continuous except for changing filters and emptying the cyclone separator. Samples were collected daily (20–28 hours) except for one 3-hour sample, four 2-day samples, and one 3-day sample (indicated by the width of the bar graphs in fig. 2).

After each collection the dust in the cyclone separator was removed with 100 ml. each of distilled wash and rinse water. Ten percent (20 ml. of the combined wash and rinse water) was filtered through a cellulose acetate membrane filter (*B*) for counting and autoradiographing the samples collected through March 22 and the sample collected on April 3. The filtrate, containing the soluble activity from the primary fraction, was evaporated to dryness in an aluminum dish (2 inches in diameter) and similarly counted in an internal counter. Twenty-milliliter aliquots of the primary fraction of all other samples were evaporated in dishes without filtration and counted, but no autoradiographs were made. Repeat counts of some samples were made to establish radioactive decay rates.

For assay of the secondary fraction, a 1⅞-inch diameter circle ($\frac{1}{23}$ of the total filter area) was punched from the center of the glass fiber filter, counted in an internal proportional counter, and used for autoradiography.

The samples were held at least 4 and usually 16 or more hours after collection to permit decay of radon daughter activity ($T_{1/2}=38$ minutes) before the beta-gamma mixed fission product activity was counted in an internal gas-flow proportional counter having an efficiency of 50 percent (*3–5*). The measured activity of a sample was corrected for decay to the midpoint of the collection time from decay data or by the application of the exponential decay factor of -1.2 for fission-product activity formed on the most probable test date.

One to four days after collection, six membrane or glass-fiber filter samples were glued face up on a 5- x 7-inch sheet of paper. The sheet was then inverted on top of the emulsion of an X-ray film (*C*). A sponge rubber pad mounted on a 5- x 7-inch piece of plywood was placed on the inverted paper and weighted with a 16-lb. brick. The film was usually exposed for 20 to 28 hours before development for normal contrast with standard X-ray chemicals, according to the manufacturer's directions.

A calibration of the optical density developed on X-ray film from exposure to fission-product particles was made by using an isolated radioactive particle in the sample collected April 5, 1955. This particle of measured beta activity was used in a series of 8 exposures ranging from 5 minutes to 19 hours and in distance of particle to emulsion from 40 to 500 microns. The density (transmission) profile of a particle autoradiograph was determined with the aid of a microphotometer which viewed a film area 46 microns in diameter by means of magnification (1 : 10) and an aperture mask on the photocell.

Photometer readings were taken progressively from the center of the irradiated area outward until the density faded into background fog. From these readings the radiation exposures at the points of measurement were calculated (*6*) and used to equate the total radiation exposure received by the film. Since the radioactivity and length of exposure were known, a proportionality constant could then be calculated for each of the eight calibration exposures. The average of the eight proportionality constants was used to calculate the radioactivity of air particulates from their autoradiographs. The calculated activity of a radioactive particle was then extrapolated from decay data to the activity at the midpoint of the collection time.

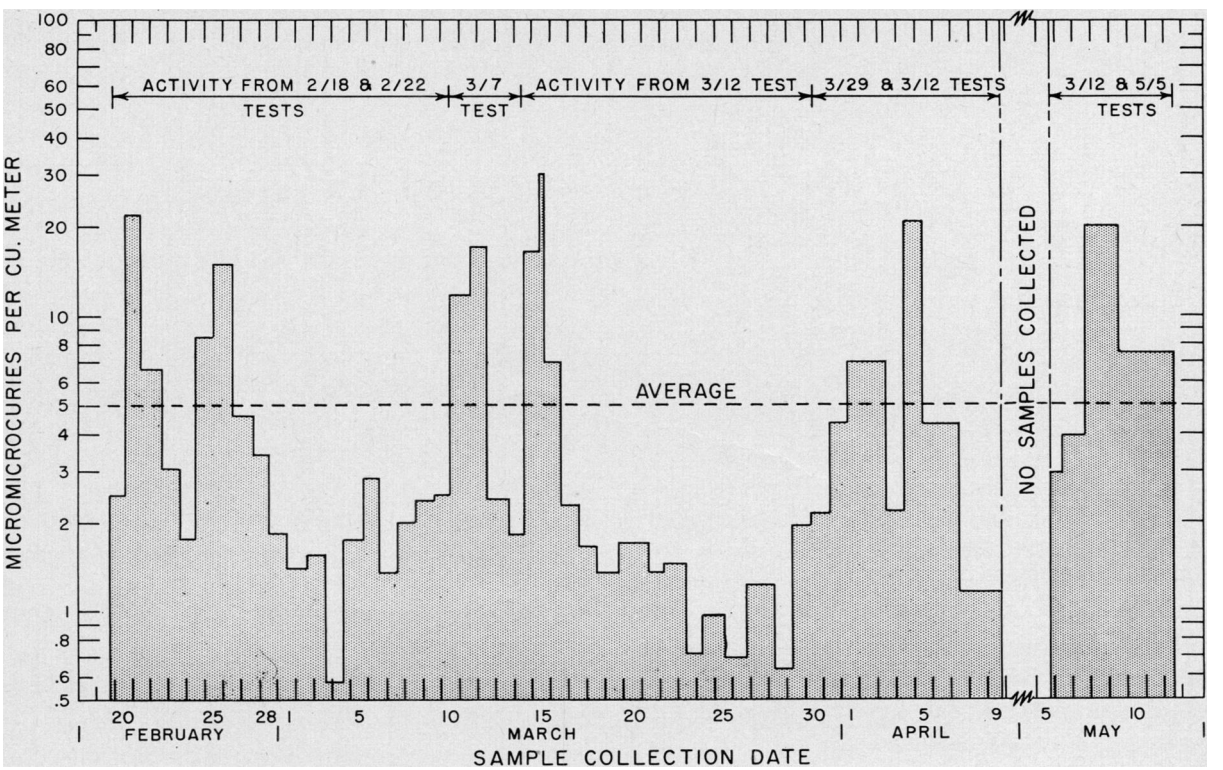
Autoradiographs of the more active particles, those which had an activity at mid-collection time greater than about 5 micromicrocuries ($\mu\mu$c.), were measured in this manner.

The minimum detectable activity by autoradiographic technique varies roughly as the square of the distance between the particle and the film emulsion. For a particle exposed for 24 hours and spaced 100 microns from the emulsion, the minimum detectable activity was estimated to be roughly 1 $\mu\mu$c. All particles having an estimated radioactivity of less than 5 $\mu\mu$c. were counted under a low-power microscope, and each was assumed to have an average activity of 2.3 $\mu\mu$c.

Gross Beta Activity

The gross beta radioactivity of particulate matter in each air sample is shown in figure

Figure 2. Gross beta activity of atmospheric particulates, Cincinnati, 1955.



2. (Gross beta activity is counted at an efficiency of 50 percent and includes gamma activity which is counted at an efficiency of 1 to 3 percent.) The lower level of beta activity found in 5 daily samples collected February 19 or later and the 2 collections made a few days before that date (not shown in the chart) ranged from 0.3 to 1 $\mu\mu$c. per cubic meter (1 $\mu\mu$c./m.^{3}=10^{-12} μc./ml.). The remaining 44 samples had an activity between 1 and 30 $\mu\mu$c. per cubic meter. Eight samples had an activity exceeding 10 $\mu\mu$c. per cubic meter. The average activity of all samples was approximately 5 $\mu\mu$c. per cubic meter.

The highest levels of activity observed in this study appeared 2 to 4 days following the reported detonation of nuclear devices in Nevada. Decay data of the air particulate samples and some rain samples were used to identify the radioactivity with specific nuclear weapons tests, as shown in figure 2.

The beta activity of particles in the primary fraction ranged from 0.08 to 17 $\mu\mu$c. per cubic meter, with an average of 1.7. The minimum value was found in the sample collected February 17. The activity of this sample had a half-life of several months. The maximum activity of a single particle was 1,700 $\mu\mu$c., found in the April 5 sample. This was the particle used for calibrating the autoradiographic technique.

Of the average activity of 5 $\mu\mu$c. per cubic meter in air particulates, an average of 1.7 $\mu\mu$c., or 34 percent of the total, was found in the primary fraction. Based on 28 samples available for comparison, from 1 to 41 percent, or an average of 16 percent, of the activity in the primary fraction was dissolved in the water used to remove the particles from the cyclone separator.

Autoradiographic Assay

Both the primary and secondary fractions of most of the samples collected from February 21 through March 19 and the sample collected on April 3 were autoradiographed after separating the water-soluble activity in the primary fraction. The autoradiographs revealed 106 active particles which were measured by photometry and 2,084 particles detected by microscopic examination.

The number of radioactive particles of variable beta activity represented an extremely small percentage of the total particles in the air, but, considering that a single person inhales 15 to 20 cubic meters of air daily, the particle radioactivity may have health significance. The variation in the number of radioactive particles in the 28 samples assayed by the autoradiographic technique is presented in figure 3. Excluding the samples of February 23 and March 1, for which only the primary fraction particles are given, the bar graph shows that all but 5 of 26 samples contained more than 1 radioactive particle per 10 cubic meters. In 5 samples there were from 16 to 82 radioactive particles per 10 cubic meters, as compared with a 26-sample average of 6.7 and a median of 4.

The beta activity of these particles ranged from an estimated minimum of 1 $\mu\mu$c. for detection by a 24-hour film exposure up to 1,700 $\mu\mu$c. for the most active particle. Three-day film exposures were also made on the samples collected February 21 and 22. About 3 times

Figure 3. Frequency of radioactive particles, Cincinnati, 1955.

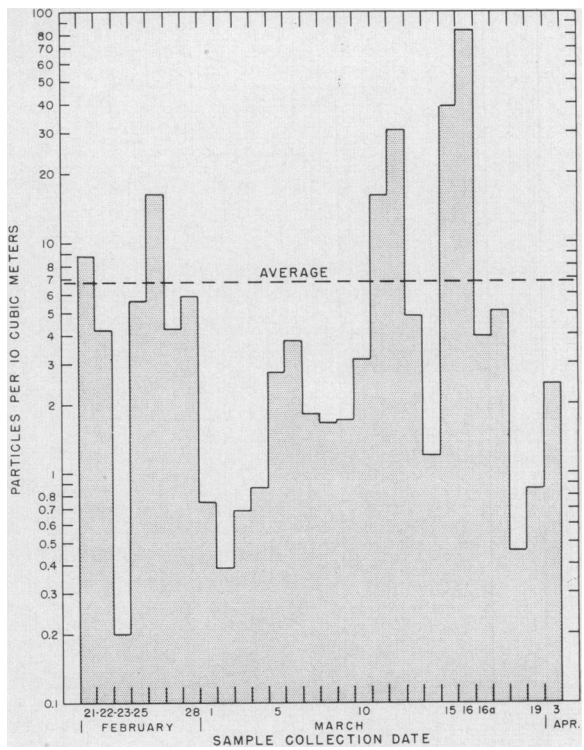
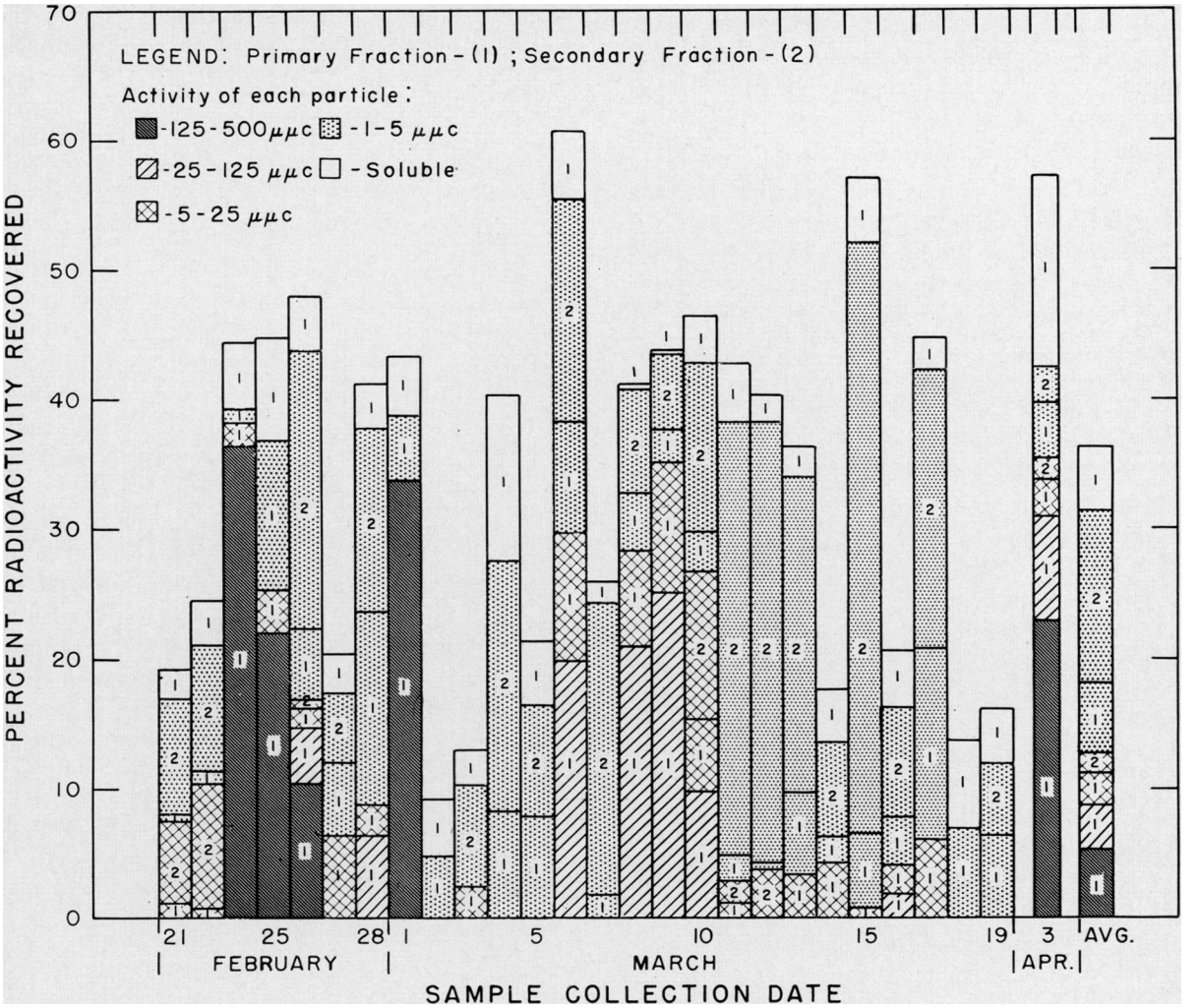


Figure 4. Radioactivity of particles as measured by autoradiographs, Cincinnati, 1955.



as many radioactive particles appeared on the 3-day autoradiographs as on the corresponding 24-hour film exposures. Thus, prolonged film exposure reveals a preponderance of particles with less than 1 μμc. of activity. It will be shown (fig. 4 and the table) that a large percentage of the total beta activity is not accounted for by the autoradiographic technique. It is likely that an appreciable part of the low-level activity from discrete particles merely increases the background fog of a 24-hour film exposure.

The particle with an estimated radioactivity of 1,700 μμc. was found in 10 percent of the primary fraction of the 24-hour sample collected at noon April 5. Subsequent autoradiographs of the entire primary sample did not reveal any more particles of activity exceeding 500 μμc. The 1,700-μμc. particle was isolated from the mass of nonradioactive particles in the sample. It was a reddish, semitransparent, nearly spherical particle having a diameter of 7.5 microns. Its decay and absorption data were characteristic of 8-day-old mixed fission product.

Distribution and Frequency

On the basis of estimates obtained either photometrically or by microscopic examination, the particles were grouped into five ranges of beta activity. The percentage distribution of

activity in each fraction and in each radioactivity range as related to the total radioactivity of each daily sample is shown in figure 4. Particles containing 125–500 μμc. were found only in the primary fraction of five samples. It is estimated that 10 to 37 percent of the total activity of these 5 individual samples resided in the "hot" particles. The samples collected March 8 and 9 each had particles with activities of 25 to 125 μμc., which accounted for 20–25 percent of their radioactivity and were recovered from the primary fraction. A considerable portion of particles in the primary fraction also had a radioactivity of 1 to 25 μμc.

The activity of particles in the secondary fraction was usually less than 5 μμc. per particle, but 6 samples had secondary-fraction particles having an activity of 5 to 25 μμc. Figure 4 shows that at most only 61 percent of the total radioactivity of a sample was observed by the autoradiographic technique. This may be explained as follows: (*a*) the secondary fraction of three samples was not autoradiographed; (*b*) particles having an activity of less than about 1 μμc. contributed only background fog to the X-ray films and this activity could not be measured; (*c*) the assumed average activity of 2.3 μμc. for particles having an activity of 1–5 μμc. each may be an underestimate; and (*d*) the autoradiographic technique is at best a rough estimate of particle radioactivity.

Occurrence of particles of given radioactivity, Cincinnati, 1955

Particle activity range, μμc.	Primary fraction		Secondary fraction	
	Maximum	Average	Maximum	Average
	Number of radioactive particles per 10 cubic meters of air			
1–5	8. 35	1. 17	73. 5	5. 2
5–25	. 38	. 12	1. 5	. 16
25–125	. 13	. 025	0	--------
125–500	. 13	. 015	0	--------
1,700	[1] 1. 06	--------	0	--------
	Percentage of fraction radioactivity in discrete particles [2]			
1–500	89	57	51	23

[1] One particle in 10 percent of sample had a diameter of 7.5 microns and an activity of 1,700 μμc.

[2] Based on autoradiographic technique employing 24-hour film exposure and, in the case of the primary fraction, its soluble activity which varied from 1 to 41 or an average of 16 percent of the total primary activity.

The results are further summarized in the table to show the frequency of occurrence of radioactive particles in the atmosphere tested. The most radioactive particle found (1,700 μμc.) had a diameter of only 7.5 microns. It appeared in the primary fraction and theoretically would occur at a frequency of 6 particles per 1,000 cubic meters of air, whereas a maximum of 13 and an average of 1.5 particles having an activity of 125–500 μμc. would occur per 1,000 cubic meters of air. The great preponderance of radioactive particles were found to have low activity levels of 1–5 μμc.; they were much more abundant in the secondary fraction. There were no particles in the secondary fraction which had an activity exceeding 25 μμc.

Of the radioactivity in each fraction an average of 57 percent was accounted for by autoradiography in the primary fraction, and an average of 23 percent was accounted for by autoradiography in the secondary fraction. Considering the radioactivity of both fractions, a maximum of 61, a minimum of 9, and an average of 36 percent of the total radioactivity was accountable by the autoradiographic technique.

Discussion

The maximum concentration of gross beta activity of 30 μμc. per cubic meter of air in the samples collected in this study was far below the 1,000 μμc. per cubic meter of air listed by the National Committee on Radiation Protection and Measurement as the provisional level for "permissible concentration of unknown mixtures of radioisotopes in the air . . . beyond the areas that are under the control of the installation responsible for the contamination." This value set by the committee is believed "to be safe for exposure to any of the radioisotopes for periods of a few months" (*7*).

About one-third of the total radioactivity of the air particulates collected in Cincinnati, that found in the primary fraction, would probably be retained in the upper respiratory system of

man. The rest would penetrate into the alveoli of the lungs. Presumably 50 percent might lodge in lung tissue for a variable time. Most of the particles in this secondary fraction had an activity of less than 5 $\mu\mu$c. per particle.

Some of the particles found in the primary fraction, however, might reach lung tissue. Some of these particles had much higher radioactivity than the secondary fraction particles. One particle with an activity of 1,700 $\mu\mu$c. had a diameter of 7.5 microns. Its size was such that if the particle were inhaled it would have a finite chance of reaching the alveoli. Once there, the particle would probably remain to cause considerable radiation damage to local lung tissue. When inhaled the smaller particles are more likely to reach lung tissue, but they may be eliminated by several mechanisms (*8–11*).

Summary

Radioactive particles found in air samples collected in Cincinnati following nuclear detonations in Nevada were separated into two size-fractions representative of particles retained by the upper and the lower respiratory system of man. Air sampling was continuous from mid-February until early April and again for 1 week in May 1955.

Measurements by autoradiographs and gross internal proportional counting indicated that:

1. The maximum, minimum, and average concentrations of beta activity in 47 samples collected between February 16 and April 9 and 4 samples collected between May 5 and May 12 were 30, 0.35, and 5 $\mu\mu$c. per cubic meter of air, respectively. For comparison, the maximum permissible level for unknown radioisotopes beyond the control area is 1,000 $\mu\mu$c. per cubic meter of air.

2. The average number of radioactive particles per 10 cubic meters of air was 6.7 in 26 samples collected between February 21 and March 19 and on April 3. The weighted average beta activity of these samples was 37 $\mu\mu$c. per 100 cubic meters of air. One sample contained as many as 80 radioactive particles and a beta activity of 300 $\mu\mu$c. in 10 cubic meters of air.

3. The more radioactive particles were collected by a cyclone separator, which simulates capture of particles by the nasal passages and bronchial tubes. Generally these particles would be rapidly eliminated from the human system. In roughly half of the samples tested, these particles had a gross beta activity of 25 to 500 $\mu\mu$c. and one isolated particle of 7.5 micron diameter had an activity of 1,700 $\mu\mu$c.

4. The activity of particles caught on the filter, representing material which would penetrate to the lung alveoli, was usually less than 5 $\mu\mu$c. and did not exceed 25 $\mu\mu$c. per particle.

EQUIPMENT REFERENCES

(*A*) Aerotec tube, design 2, Thermix Corp., New York, N. Y.

(B) Millipore filter, type HA, Millipore Filter Corp., Watertown, Mass.

(*C*) Blue Brand X-ray film, Eastman Kodak Co., Rochester, N. Y.

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Regional Medical Directors Reassigned

The Public Health Service has announced a series of changes among medical directors assigned to the regional offices of the Department of Health, Education, and Welfare. The changes will become effective July 1, 1958.

Dr. Francis J. Weber, medical director of Region 8, with headquarters in Denver, Colo., has been appointed chief of the Service's newly created Division of Radiological Health in Washington, D. C.

Dr. Michael Pescor, present medical director of Region 7, with headquarters in Dallas, Tex., will become medical director of the Denver office.

Dr. Richard Boyd, now medical director of Regions 1 and 2, stationed in New York City, will assume the same position in the Dallas office.

Dr. Harald M. Graning, medical director of Region 5, in Chicago, will move into the New York City position.

Dr. Arthur B. Price, at present on loan from the Public Health Service to the Bureau of Old-Age and Survivors Insurance, Social Security Administration in Baltimore, will become the medical director of the Chicago office.

The medical directors of other regions are: Dr. Eugene Gillis, Region 3, Charlottesville, Va.; Dr. Will H. Aufranc, Region 4, Atlanta, Ga.; Dr. Lewis H. Hoyle, Region 5, Kansas City, Mo.; and Dr. Charles F. Blankenship, Region 9, San Francisco.

The medical directors in the regional offices direct Public Health Service programs of grants and technical assistance and other relationships with the States and Territories that comprise each region.