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AN EVALUATION OF THE PULMONARY RADIATION DOSAGE FROM RADON AND ITS DAUGHTER PRODUCTS

by:

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AN EVALUATION OF THE PULMONARY RADIATION DOSAGE FROM RADON AND ITS DAUGHTER PRODUCTS

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Calculations and experimental results with the dog both indicate that the upper respiratory passageways in the lung receive the largest pulmonary radiation dosages from the deposited daughter products.

The sampling of an atmosphere for the radon daughter products is discussed in detail.

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AN EVALUATION OF THE PULMONARY RADIATION DOSAGE FROM RADON AND ITS DAUGHTER PRODUCTS*

INTRODUCTION

1.1 Purpose.

The extent of the radioactive matter deposited in the lungs by humans breathing in air contaminated with radon was recognized only recently (1). The radon atoms in the air are the source of short-lived polonium, lead, and bismuth daughter atoms. These daughter atoms generally become attached to dust particles and other solid matter present in the environment where they are produced. The evaluation of the radiation hazard to the lungs of humans breathing in air containing radon thus involves not only an evaluation of the dosage from the radon gas atoms but of the dosage from the radioactive aerosol produced by the radon daughter products. Very little information is available on the nature of the radioactive aerosols produced by radon under different environmental conditions and the extent of the retention of these aerosols by the human respiratory tract.

The investigations reported here were concerned with the determination of the radiation dosages imparted to the lungs of rats, dogs, and man breathing air containing radon and its daughter products. Measurements were made of the daughter product radioactivity deposited in the lungs of rats and dogs after they had been breathing for several hours in an exposure chamber containing atmospheric air to which radon had been added. Values were obtained for the per cent

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* The material in this paper was submitted to the University of Rochester in partial fulfillment of the requirements for the Doctor of Philosophy degree in Biophysics.

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deposition of the inhaled daughter products in the lungs of man breathing from mixtures of radon with both uncleaned atmospheric air and filtered atmospheric air. An attempt was made to evaluate the radiation hazard from radon and its daughter products to the different regions of the human respiratory tree for various sizes of daughter product carriers. Several procedures for sampling a region for the air-borne daughter product activity were devised and tested.

The Subcommittee on Permissible Internal Dose of the National Committee on Radiation Protection (2) has recommended on the basis of data available to 1951, that the maximum permissible concentration of radon in equilibrium with its daughter products in the air be 10^{-11} curies per liter for continuous exposure. For a forty-hour work week the maximum permissible concentration is then 4.2×10^{-11} curies per liter. The median measured radon concentration in uranium mines in the Colorado Plateau is 3.1×10^{-9} curies per liter (3). The radon levels in many of the industrial installations where radium containing substances are handled cannot be practically reduced much below 10^{-10} curies per liter. These levels make radon a major source of human exposure to radioactivity at the present time. It was with the purpose of furthering the understanding of the extent and control of the hazard from breathing in an atmosphere containing radon that this investigation was undertaken.

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1.2 Background.

In 1940 Evans and Goodman (4) calculated the dosage delivered to the human lung by the alpha particles from radon itself and from the daughter products of the radon molecules decaying in the lung. On the basis of their calculation, a concentration of 10^{-9} curies per liter of radon breathed forty hours per week led to a dosage, averaged over the lung, of 5.03 mrep per week, or 101 mrem per week of alpha radiation. In another calculation Evans (5) assumed that the important radiation dosage was to cells lining the bronchial passageways. For a forty-hour week and a radon level of 10^{-9} curies per liter, this type of calculation gave a dosage of 172 mrem. Evans and Goodman calculated for a radon level which they considered characteristic of the measured radon levels in the radium mines of Schneeberg, Germany and Joachimsthal, Czechoslovakia.

The death rate among the miners of Schneeberg and Joachimsthal from pulmonary diseases, especially lung cancer, was very high. When the Czechoslovakian Ministry of Health became aware of the existence of lung cancer among the miners in Joachimsthal it undertook post-mortem examinations on the miners and pensioners. Between 1929 and 1938, 89 miners and pensioners died. Of the 89 deaths, 47 were from cancer. In 43 of the 47 cases, the primary tumor was found in the respiratory organs; 42 were considered as intrathoracic tumors and one as carcinoma

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of the larynx. Post-mortem examinations of 30 of the non-cancerous cases revealed that 24 of the men had died of lung diseases. Peller (6) tabulated the diagnoses and the ages of death of the miners and pensioners. His table is reproduced in Table 1.

Table 1.

Diagnosis of Death and Age at Death of Joachimsthal Miners from S. Peller, Human Biology 11, 130(1939)

Age at death	Examined post-mortem 1929-1930, 1933-1938		Not examined post-mortem 1929-1932	
	Cancer	Non-Cancer	Cancer	Non-Cancer
-29		1		
30-34		3	1	3
35-39	2	10	4	1
40-44	9	10	3	3
45-49	8	4	3	
50-54	8		2	
55-59	1		3	2
60-64	2			
65-69		1		
70-74				1
75-79			1	2
?		1		
	30	30	17	12

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Evans and Goodman (4), assuming radon was the carcinogenic agent, summarized the published data on the radon content of the air in various mines of the Schneeberg and Joachimsthal districts. Six samples were taken in each of two Joachimsthal mines between 1924 and 1925. The radon concentrations per liter of air, in units of 10^{-9} curies, ranged from

0.3 to 1.8 with an average of 0.8 for mine A;
2.3 to 8.8 with an average of 5 for mine B.

Evans and Goodman considered the average of the various determinations, which was 2.9×10^{-9} curies per liter, as representative of conditions in the mines. Using a safety factor of 100, they took 10^{-11} curies per liter as the safe working concentration of radon or thoron in the air.

Their tolerance figure is widely used in the radium dial painting industry. Since it is based primarily upon human exposure data, it depends for its validity upon the accuracy of the radon levels as reported for these European mines and on the accuracy of the assumption that these lung cancers were in fact radon produced. It may be noted that the mine radon values were accumulated at the time a high incidence of lung cancer was recognized, and may not, even if accurate in themselves, be representative of the effective concentrations encountered by the miners over the many years of exposure presumably necessary to produce the lung cancers.

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In 1944 Lorenz (7) reviewed the available literature on lung cancer in the European miners. He concluded that a number of environmental factors could have contributed to the cause of the lung cancers. He made a dosage calculation similar to the one made by Evans to show that the radon levels in the mines did not seem to have been high enough to have produced the lung cancers.

In 1951 Bale (1) pointed out that the dosage imparted to the lungs in air containing radon was much higher than the published calculations had indicated. He observed that these calculations had neglected the radiation dosages produced by inhaling the daughter products of radon present in the air. These daughter products become deposited largely on particulate matter in the air soon after they are produced and thus create a radioactive aerosol which remains suspended in the air. The dust is breathed in along with the radon and a fraction of the inhaled dust particles is retained in the lungs. The result is that the daughter products build up to much higher levels in the lungs than would be attained on the basis of simple equilibrium with the inhaled radon. The radiation dosage from the inhaled daughter products was calculated by Bale to be many times higher than the dosage from the radon in the lungs and the daughter products of the radon molecules decaying in the lungs.

Experimental results which confirmed these conclusions were reported independently by Cohn et al (8), and by Bale and the writer (9) in 1952. The latter work is covered in this thesis.

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High gamma activities were measured in lungs removed from rats which had been breathing air containing radon in equilibrium or near equilibrium with its daughter products. Almost all of this activity resulted from the deposition of the inhaled daughter products. The gamma activity of the lungs of rats exposed to air-radon mixtures which were well cleaned of the daughter products was only a small fraction of the activity when the daughter products were not removed.

2. REVIEW OF PUBLISHED DATA ON RADIATIONS AND RADIOACTIVE MATTER IN AN ATMOSPHERE CONTAINING RADON.

When air containing radon gas is inserted into a chamber whose walls do not adsorb radon, the radon molecules soon become distributed uniformly throughout the chamber. These radon molecules, with a 3.825 day half-life, are the source of a series of radioactive transformations. That part of the decay scheme which is relevant to the present investigation is given in Fig. 1.

2.1 Buildup of Radon Daughter Products from Fresh Radon.

Radium A, the first decay product of radon, will build up until its disintegration rate equals its production rate. It will reach 50 per cent of its final equilibrium value in 3 minutes. RaB and RaC will attain half their final equilibrium values in about 30 minutes and 60 minutes respectively (10). RaC', because of its extremely short half-life, is always in equilibrium with RaC and

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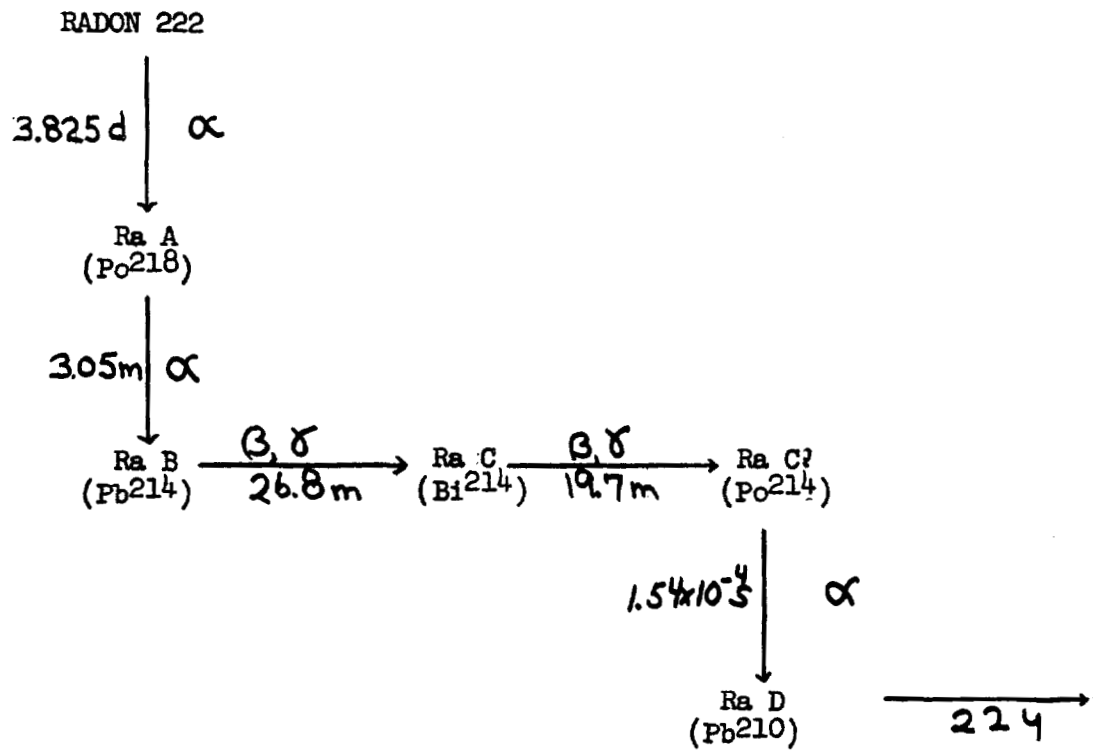

Fig. 1. Radon decay scheme.

disintegrates at the same rate. After three hours the disintegration rates of RaA, RaB, RaC, and RaC' are just slightly less than that of radon.

2.2. Properties of Radon Daughter Products; Active Deposit (11, 12).

Early investigators in the field of radioactivity observed that when any material was inserted into an atmosphere containing radon, it soon became coated with a film of radioactive matter. When the material was removed from the radon atmosphere, the activity decayed fairly rapidly, reaching a negligible amount in the period of a few hours. Because this activity seemed to collect everywhere

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that radon was present, both on the walls of the confining vessel and on any type of material in the vessel, it was called the "active deposit" of radon.

Curie reported that the radioactivity deposited on different plates inserted into a radon atmosphere was independent of the composition of the material. Glass, paper and metals were made equally radioactive.

The activity was independent of the nature and pressure of the gas (air, hydrogen, carbon dioxide) with which the radon was mixed in the enclosure.

The activity of bodies in a closed container was proportional to the free space, between 1 mm. and 3 cm., in front of them.

Washing a metallic surface containing the active deposit with water did not remove much of the active deposit.

Rutherford reported that the active deposit of radon in air contained between oppositely charged plates concentrated mainly on the negative electrode. Under ordinary experimental conditions about 5 per cent appeared on the anode. The small activity which appeared on the anode in a very strong field was shown to be due to diffusion of uncharged atoms of active deposit (13). Rutherford obtained the value of 1.3 $cm.sec.^{-1}$ $volt^{-1}$ $cm.^{-1}$ for the mobility of the radon daughters, a value near that of positive ions created in air by x-rays.

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2.3. Diffusion of Active Deposit.

Curie studied the propagation of the active deposit by diffusion. The experiments were performed in a thirteen liter jar. The gas was carefully dried. The activity deposited on a series of planes placed at various distances apart was studied as a function of the radon concentration. It was observed that the activity increased as the planes were separated to a maximum limiting distance which depended on the concentration. For a radon concentration of 3×10^{-8} curies per liter, this limiting distance was 38 mm. For 75x this concentration it was 25 mm., and for 375x the concentration it was 19 mm.

Chamie (14), working with the thorium daughter products, found that the collected quantity of ThC" on a plate a short distance from the ThC plate source was independent of the distance between the source and collector as long as this was smaller than the range of nuclei in the gas (0.13 mm.). When this distance exceeded the range but was less than 2-3 mm. of air, the efficiency of collection decreased in inverse proportion to the distance.

Langevin (15), using these experimental results, made a mathematical analysis of the diffusion and deposition of the thorium daughter products. He interpreted the results in terms of competitive collection by the emitting and collecting surfaces. He concluded that the nuclei diffused in the intervening space like molecules of a gas but were adsorbed on the surfaces with high efficiency. The

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steady state concentration of activity in the space remained small.

In further experiments along the same lines, Chamie and Tsien (16) showed that at larger distances than 5-25 mm., the apparent rate of diffusion to the walls was much reduced and the steady state concentration of radioactivity in the space could not be small any more. This was explained as being due to the adsorption of diffusing recoil atoms on dust.

2.4. Gravity Effect.

Curie observed a gravity effect when the radon concentration was high. Measuring the activity on the surfaces of two parallel planes which were facing each other, she found that surfaces looking up in a jar picked up more activity than the surfaces looking down. Filtering the air did not affect the results, so apparently dust particles were not involved. However, water vapor appeared to be necessary for the production of the effect.

The importance of the gravity effect grew with the concentration of the radon and with the distance of the parallel plates. It did not appear for faces 2 mm. apart but appeared at distances of 1 cm. to 3 cm. It didn't appear under 2 or 3 cm. Hg. so the pressure seemed to be important. She concluded that in the presence of traces of water vapor there were agglomerations which carried the active deposit and which were large enough to undergo the action of gravity.

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2.5. Association of Gravity Effect with Clustering.

The phenomenon of radioactive clustering was studied by Chamie (17) and by Harrington (18, 19) and co-workers. Chamie directed air containing radon gas against a photographic plate and noticed stars in the developed image.

Harrington made an extensive study of clustering. He used a small tube, 6 cm. long, filled with 15 mc. of radon and air at atmospheric pressure. He studied the activity along the tube before and after centrifuging. Without centrifuging, the activity was fairly unifrom. After centrifuging under a field of 500 g , he found that most of the activity appeared at the end of the tube, thus showing a gravity effect. The magnitude of the effect produced by centrifuging increased with the partial pressure of radon and of water vapor and with the total pressure and depended somewhat on the shape of the tube. Filtered air was used.

He was also able to observe the clusters with the ultramicroscope. He found that they were quickly swept to one side in an electric field. Most of the aggregates carried a positive charge, but many also carried a negative charge. The mobilities of the molecular aggregates fell between 10^{-4} and 6×10^{-4} cm. sec.$^{-1}$ volt^{-1} cm.$^{-1}$. This was of the same order of magnitude as those reported by Langevin for large ions produced by x-rays.

Martin, Baylor and Clark (20) studied cluster formation by the thorium degradation products with the electron microscope,

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collecting the degradation products on a collodium film supported on a 200 mesh copper screen. They reported breaking up of the clusters in time, indicating that the emission of alpha particles was sufficient to break up the aggregate.

2.6. Distribution of Daughter Product Activity in a Flask.

Maxwell, Henri and Peterson (21) reported experiments on the distribution of the radon decay products in a gas filled spherical vessel. Some of their results are given in Table 2. They observed a gravity effect.

Table 2.

Distribution of Daughter Product Activity in a Flask from Maxwell et al., J. Chem. Phys. 18, 179 (1950)

Radius of flask mm.	Pressure of gas mm. Hg. C_6H_6	Pressure of gas mm. Hg. CO_2	Radon mc.	Per cent of total daughter product activity in flask deposited on walls: % RaC on wall	Per cent of total daughter product activity in flask deposited on walls: % RaA+RaB on wall
90	3		15.2	96.4	91.5
90	80		14.5	52	41
90	80		310	78	64
64		17	20	99	97
64		640	24	94	90

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2.7. Experiments on Air-borne Daughter Product Activity in a Room.

Harley (22) made an extensive study of the air-borne radon daughter products in a 1000 ft.^{3} room. His radon concentration was about 2.9×10^{-11} curies per liter. He used various filters to remove the daughter products from the air and then measured the alpha activity deposited on the filter paper.

In one experiment, air in the room was made relatively dust free by scrubbing the room down and cleaning the air as thoroughly as possible by filtration. Under these conditions the alpha activity measured on a millipore molecular filter was about 40 per cent of the maximum possible activity. Harley also studied the radon daughter product activity in a high dust atmosphere. The atmosphere was obtained by spraying a fine mist of aqueous sodium chloride into a dryer placed in the sampling room. Under these conditions he obtained 75 per cent of the maximum possible daughter product activity. His experiments did not determine whether the daughter products were in the air but not retained by the filter, or whether they had been removed by the walls of the chamber.

Charmichael and Tunnicliffe (23) also measured the radon daughter product activity suspended in room air. They removed the decay products with an electrostatic precipitator. They reported that they removed 80 per cent of the daughter product activity produced by the radon per unit volume of air sampled.

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2.8. Distribution of Radon Daughter Products on Atmospheric Dust.

About 96 per cent of the radioactivity found on particulate matter in the atmosphere is produced by the daughter products of the radon in the air. Wilkening (24) measured the mobility of the carriers of the radioactivity with an electrostatic precipitator and obtained the particle size with the aid of Stoke's law. He found that 90 per cent of the natural radioactivity he removed from the air was attached to particles that had effective diameters less than 0.5 mu. Most of this fraction resided on particles in the size range 0.001 mu. to 0.04 mu. The major group at 0.018 mu. agreed well with that of the so-called "intermediate" atmospheric ions having mobilities of 0.02 $cm.sec.^{-1}volt^{-1}cm.^{-1}$.

2.9. Measurements of Radon and Radon Daughter Products in American Uranium Mines.

Measurements of the radon and daughter product levels in the air of the uranium mines of the Colorado Plateau have been made under the direction of Holaday since 1950 (3, 25). The radon levels vary considerably with a median value of 3.1×10^{-9} curies per liter. The corresponding daughter product concentrations also vary over a wide range with most of the values between 30 per cent and 80 per cent of the concentration which would be in equilibrium with the radon in the air.

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3. CALCULATED RADON DAUGHTER PRODUCT DECAY CURVES.

3.1. Decay Curves for Very Short Sampling Time.

We can obtain the relative distributions of RaA, RaB, and RaC-C' in air containing radon by removing the daughter products from the air and analyzing their decay curve. A simple method of obtaining a daughter product sample is to draw the air very quickly through a suitable filter. Alpha, beta, or gamma counts can then be made on the daughter products removed by the filter. The shape of the decay curve of this rapidly taken or "grab" sample is more sensitive to variations in the relative concentrations of RaA, RaB, and RaC-C' in the sampled air than the shape of the decay curve of an air sample obtained over an extended period of time.

The theoretical decay curves are obtained by solving the series of first order differential equations relating production and decay of each member in the decay series and inserting the appropriate initial conditions. Using the data in Table 3 one obtains the following equations for the disintegration rates of RaA, RaB and RaC-C'.

For RaA. $\lambda_2 N_2 = \lambda_2 N_2^\circ e^{-\lambda_2 t}$

For RaB. $\lambda_3 N_3 = -0.128\,\lambda_2 N_2^\circ e^{-\lambda_2 t} + (\lambda_3 N_3^\circ + 0.128\,\lambda_2 N_2^\circ) e^{-\lambda_3 t}$

For RaC-C' $\lambda_4 N_4 = 0.023\,\lambda_2 N_2^\circ e^{-\lambda_2 t} + (3.775\,\lambda_3 N_3^\circ + 0.485\,\lambda_2 N_2^\circ) e^{-\lambda_3 t}$
$+ (\lambda_4 N_4^\circ - 3.775\,\lambda_3 N_3^\circ - 0.508\,\lambda_2 N_2^\circ) e^{-\lambda_4 t}$

λ = disintegration constant; N = number of atoms

$\lambda_2 N_2^\circ, \lambda_3 N_3^\circ, \lambda_4 N_4^\circ$ = initial disintegration rates of RaA, RaB, RaC-C' respectively.

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Table 3.

Radiations from Radon and Its Daughter Products

Element	Common name	1/2 life	Disint. constant m^{-1}	Particle radiation	Energy Mev	Range in water(27) microns	Range in air(26) cm.
Rn^{222}	Radon	3.825 d	12.6×10^{-5}	α	5.49	39	4
Po^{218}	RaA	3.05 m	0.2273	α	5.998	42	4.3
Pb^{214}	RaB	26.8 m	0.02586	β	0.65		
Bi^{214}	RaC	19.7 m	0.03519	β	3.15		
Po^{214}	RaC'	1.54×10^{-4}s		α	7.68	68	6.9
Pb^{210}	RaD	22 y					
...							

Gamma Radiation from RaB and RaC[28]

RaB		RaC	
Photon energy Mev	Average quanta per Rn α[a]	Photon energy Mev	Average quanta per Rn α[a]
0.241	0.115	0.607	0.658
0.294	0.258	0.766	0.065
0.350	0.450	0.933	0.067
		1.120	0.206
		1.238	0.063
		1.379	0.064
		1.761	0.258
		2.198	0.074
	0.823		1.455

[a] Assuming equilibrium between radon and its daughters.

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The decay curves of RaA and RaC' have been plotted in Fig. 2 for the case where the daughter products are initially in equilibrium. The RaC' curve has been broken down into curves which give the contributions from the decay of RaA, RaB, and RaC to the RaC' disintegration rate.

The initial disintegration rates of RaA, RaB, and RaC-C' sampled instantaneously from a stable atmosphere containing radon will be equal if the daughter products are not removed selectively after they are formed. Thus, the disintegration rates will be equal for radon in a stable atmosphere of infinite size. They will also be equal for a finite atmosphere containing a very high and stable dust load. The radon daughter products then become attached to the dust particles and remain in the air. In practice many of the daughters are removed from the air in a finite enclosure by depositing out on the walls. The later members of the decay series will be removed to a greater extent than the earlier ones, since their later times of decay give them a greater chance to reach the walls. Accordingly, pertinent conditions for calculation include only cases where the initial RaA disintegration rate per unit volume is equal to or greater than that of RaB. Similar restrictions apply to RaB and RaC.

Once the disintegration rates of the elements in the series are known, the activities which would be measured can be calculated from a knowledge of the radiations given off. Thus the total

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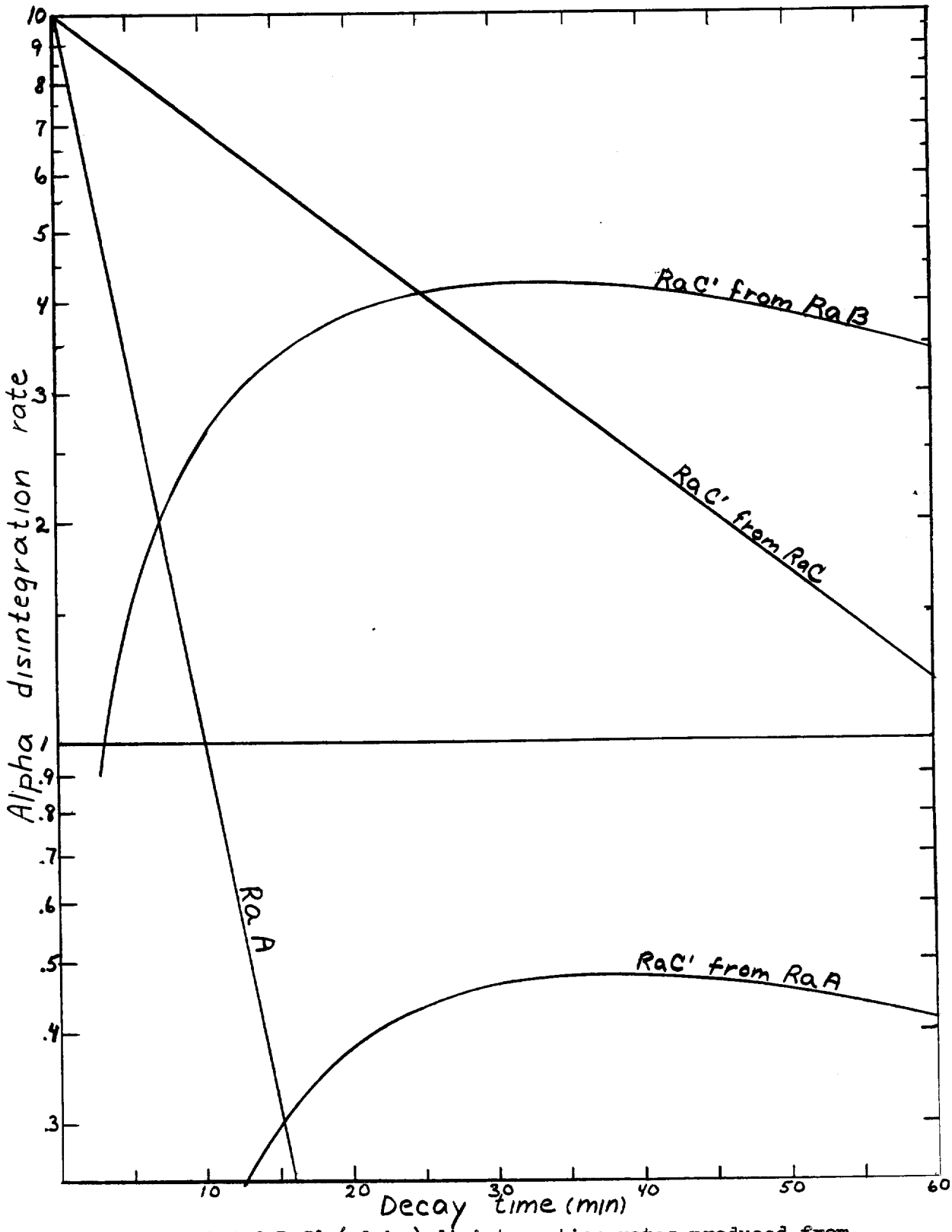

Fig. 2 RaA and RaC' (alpha) disintegration rates produced from decay of RaA, RaB and RaC, each with an initial disintegration rate of 10.

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alpha rate is the sum of the disintegration rates of RaA and RaC'; the total beta rate is the sum of the disintegration rates of RaB and RaC; and the total measured gamma disintegration rate is the sum of the disintegration rates of RaB and RaC, each multiplied by the average number of photons given off per disintegration and the efficiency of detection per photon of the specified energy.

Alpha decay curves are plotted in Fig. 3 for several different relative initial disintegration rates of RaA, RaB, and RaC-C'. They were obtained with the aid of Fig. 2. Gamma and alpha decay curves starting with RaA, RaB, and RaC-C' in equilibrium are plotted in Fig. 4.

The theoretical gamma decay curves were derived on the assumption that the soft and hard gamma radiations were detected with equal efficiency. This was a satisfactory assumption since experimental data to be reported later indicated that the detection efficiency of the counter used was not very different for the various photon energies emitted by the daughter products. It can also be shown that the shapes of the decay curves are not very sensitive to moderate variations in the detection efficiencies of the different gamma energies.

3.2. Decay Curves for Infinite Sampling Time.

We shall want to know the equilibrium buildup of the daughter products on a filter paper which samples air from a given

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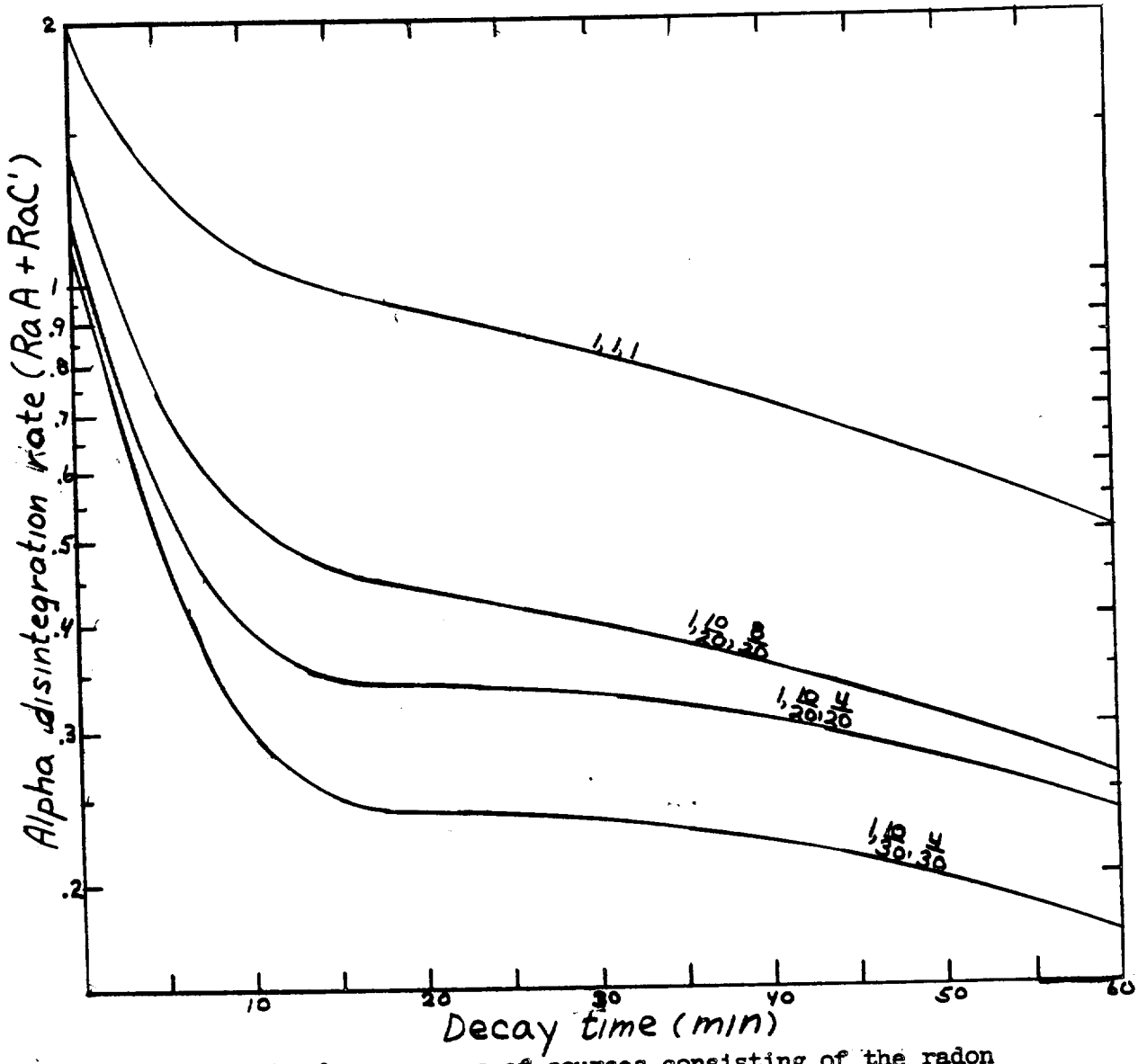

Fig. 3 Alpha decay curves of sources consisting of the radon daughter products, RaA, RaB, and RaC-C' for different initial disintegration rates of the daughter products. The first, second and third digits on each curve give the relative initial disintegration rates of RaA, RaB and RaC-C' respectively. The initial disintegration rate of RaA has been arbitrarily chosen as one in each case.

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Fig. 4. Decay curves of the radon daughter products after they are separated from a constant radon source for the case where they were all initially in equilibrium with the radon. The ordinate gives the ratio of the activity of the daughters to the activity of the radon source.

Curves A, B, and C are the decay curves for RaA, RaB, and RaC-C' respectively. Curve D is the sum of A and B. It gives the alpha disintegration rate. Curve E = 1.445B + 0.823C. It gives the ratio of the gamma disintegration rate to the disintegration rate of the radon.

Selected numerical values taken from this graph are:

Decay time min.	Activity of daughters / Activity of radon		dpm/ℓ daughters for 10^{-9} c/ℓ radon	
	α curve	γ curve	α curve	γ curve
0	2.00	2.28	4440	5060
20	.93	1.90	2065	4220
40	.71	1.36	1575	3020
60	.5	.93	1110	2065

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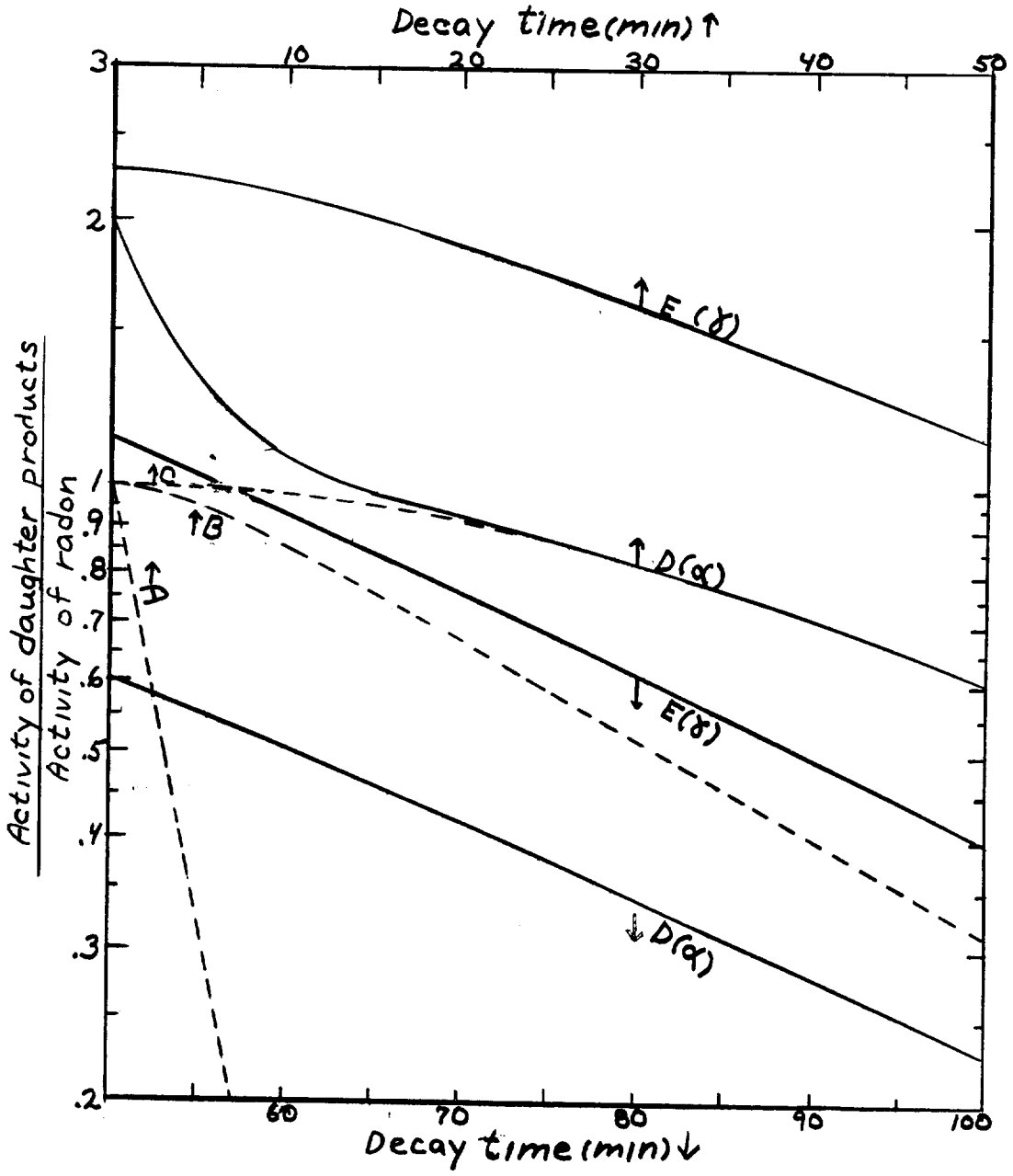

Fig. 4. (Arrow points to appropriate time scale.)

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radon atmosphere for an infinite length of time. For practical purposes, any sampling time over two hours may be considered as infinite.

When we sample through the filter paper, RaA, RaB, and RaC will build up on the filter paper till their disintegration rates equal their rates of growth. The rate of growth of any one daughter product equals the rate at which it is removed from the medium plus the rate at which it is produced from the decay of the preceding member of the series. The number of atoms per unit time produced from the preceding member is, of course, equal to the disintegration rate of the preceding member. Thus, the maximum disintegration rates attained by RaA, RaB, and RaC-C' on the filter paper are

For RaA: $\lambda_2 N_2^o = Q_2 v$

For RaB: $\lambda_3 N_3^o = Q_3 v + \lambda_2 N_2^o$

For RaC-C': $\lambda N_4^o = Q_4 v + \lambda_3 N_3^o$

λ = disintegration constant

N_2^o, N_3^o, N_4^o = final numbers of atoms of RaA, RaB, RaC respectively

Q_2, Q_3, Q_4 = daughter atoms per unit volume of RaA, RaB, RaC respectively

v = volume of air sampled per minute

The RaC' activity effectively equals the RaC activity because the RaC' activity produced on the filter paper from the

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removal of the relatively few RaC' atoms in the sampled air is negligible in comparison to the RaC' activity produced from the decay of RaC.

If Q_1 is the number of radon atoms per liter and if we assume radioactive equilibrium between radon and its daughter products, then $\lambda_1 Q_1 = \lambda_2 Q_2 = \lambda_3 Q_3 = \lambda_4 Q_4$.

For RaA: $\lambda_2 N_2^o = 5.53 \times 10^{-4}\ Q_1 v$

For RaB: $\lambda_3 N_3^o = 54.16 \times 10^{-4} Q_1 v$

For RaC: $\lambda_4 N_4^o = 89.90 \times 10^{-4} Q_1 v$

When v is in liters per minute, λN = disintegrations per minute.

The total daughter product activity reaches 97 per cent of its final value after 2-1/2 hours sampling time.

Decay curves of activity on the infinite filter paper sample, after sampling has stopped, are given in Fig. 5. They are given both in terms of $\frac{\lambda i Ni}{Q_1 v}$ and in total counts for a sampling rate of 0.1 liter per minute in a radon concentration of 10^{-9} curies per liter, assuming 100 per cent equilibrium between parent and daughters. From these curves, one can easily obtain the disintegration rates for other values of sampling rates and radon concentrations.

Curves giving the buildup of radon decay products as a function of sampling time have been plotted by Harley (22).

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Fig. 5. Decay curves of the radon daughter products after they have built up to a steady state level in a system which has filtered them with 100% efficiency from air containing radon and its daughter products in equilibrium. The decay time is measured from the end of the filtering period. The left hand ordinate gives the ratio of the activity of the daughters in the filter to the rate at which radon atoms passed through the filter. The right hand ordinate gives the number of disintegrations per minute for air containing 10^{-9} curies per liter of radon in equilibrium with its daughters flowing through the filter at the rate of 0.1 liters per minute.

The curves marked α and γ give the alpha and gamma disintegration rates respectively.

Selected numerical values taken from this graph are:

Decay time min.	$\frac{\text{Daughter activity on filter}}{\text{Rn atoms/min thru filter}} \times 10^4$		Daughter dpm on filter for 10^{-9} c/ℓ Rn, 0.1 ℓ/min	
	α	γ	α	γ
0	95.4	175.5	16800	30950
20	65.5	122.1	11530	21540
40	44.9	81.3	7950	14330
60	29.7	52.7	5230	9300

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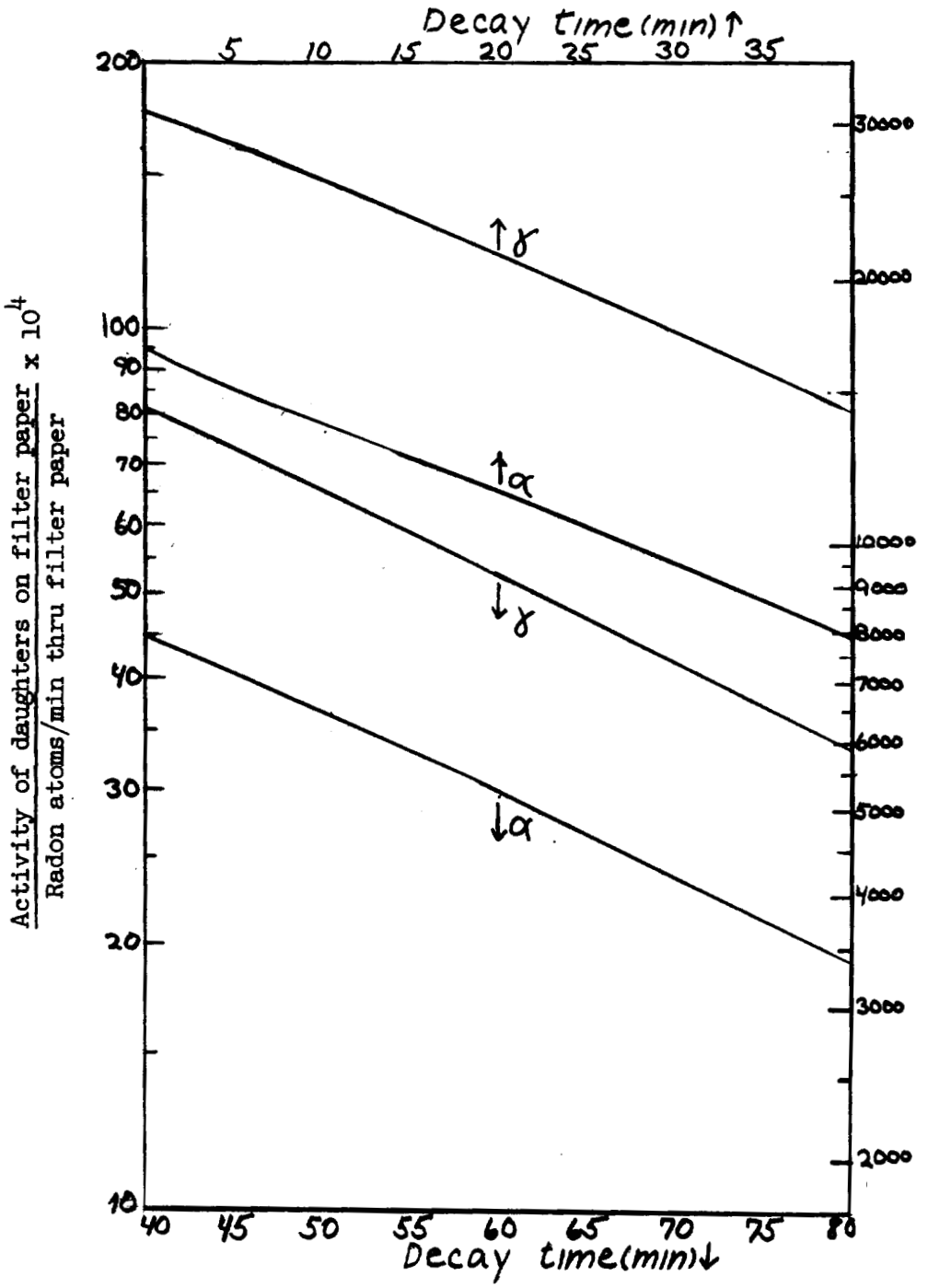

Fig. 5. (Arrow points to appropriate time scale.)

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4. EXPERIMENTAL DESIGN

4.1. Experimental Chamber.

All the sampling and animal exposure experiments were performed in a chamber 3.9 ft. by 2.9 ft. by 6.4 ft. of approximately 1800 liters capacity. The chamber was of wood construction and was painted inside and out with an enamel paint. The gaskets used on the outlets from the chamber were of neoprene.

The chamber had a large rectangular lucite window, 18 in. by 24 in. A sponge neoprene gasket sealed the lucite window to the chamber. It was found necessary to use angle iron to clamp the lucite frame to the chamber to produce a uniform pressure on the gasket.

The chamber was tested for leaks by maintaining the air pressure inside at a positive pressure of several inches of water and painting soap on the outside. The formation of bubbles pointed out the leaks, which were then sealed with glyptal. Fig. 6 is a photograph of the chamber.

4.2. Establishment of Radon-Radon Daughter Product Atmosphere in Chamber.

In experiments to study daughter product retention in an atmosphere containing "normal atmospheric dust," outside air was blown through the chamber during the night preceding the first day of a given series of experiments by means of a fan placed in front

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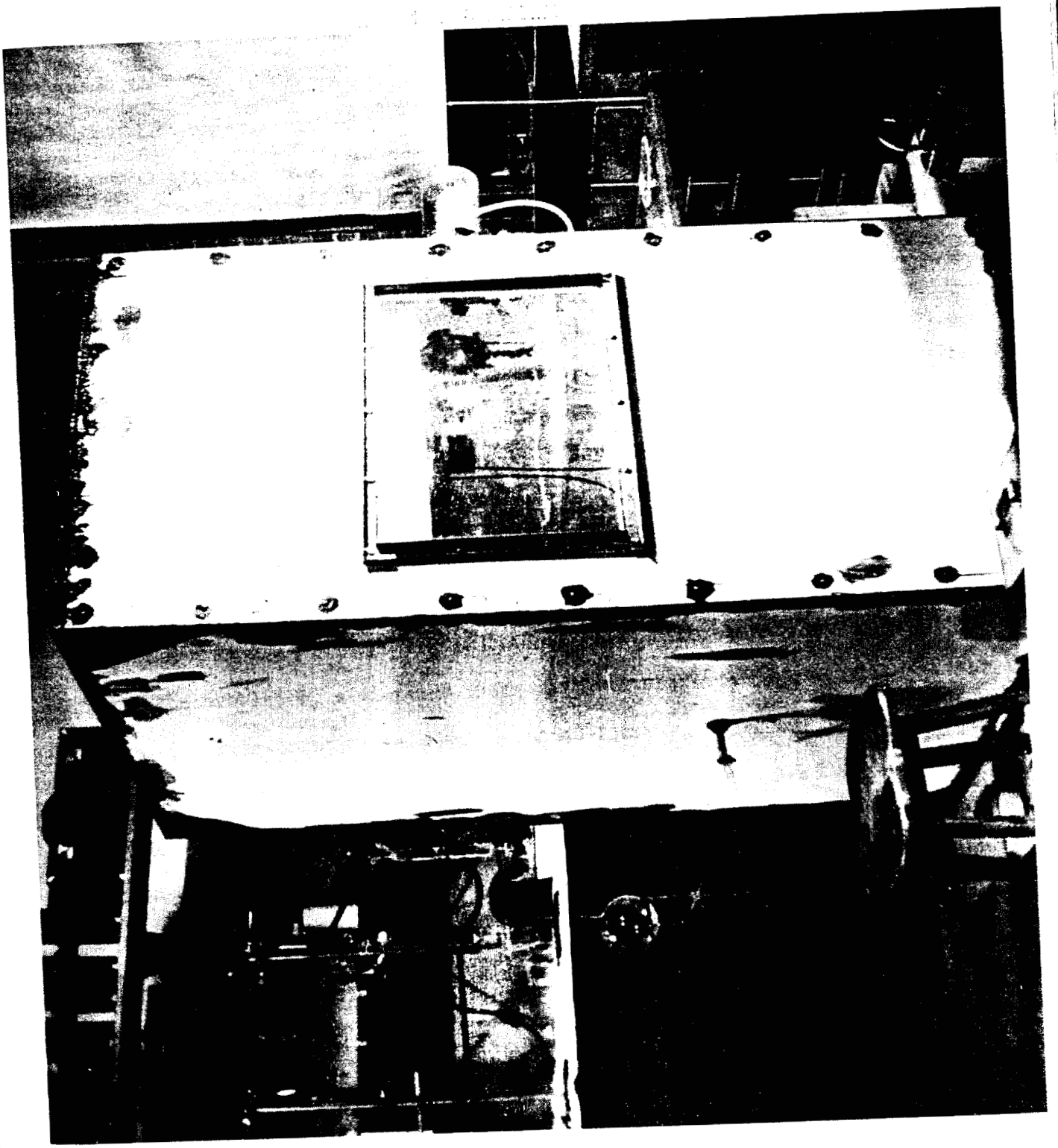
Fig. 6. Chamber used for animal exposure and sampling experiments.



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of an open window. In the morning the chamber was sealed and radon inserted.

In experiments to study daughter product deposition in filtered air, air from the compressed air-line was filtered with a millipore filter before being blown through the chamber. The air was blown through the chamber during the night preceding the first day of the experiments. In the morning the chamber was sealed and radon inserted.

A period of at least two hours was allowed for the radon daughters to come into equilibrium with the radon in the chamber before beginning the first experiment.

The radon for the chamber was collected with a deemanation apparatus similar to that described in University of Rochester Atomic Energy Project Report UR-255 (29). Air which was broken into fine bubbles by a sintered glass disk was drawn through a radium chloride solution into an evacuated liter bulb. A small length of thermometer tubing between the radium solution and the collecting flask insured very slow passage of the air through the solution. The air was passed through the solution for about ten minutes, after which the solution was disconnected from the collection flask. Room air was then flushed through the system to bring the collecting flask up to atmospheric pressure. This method removed almost all the radon produced by the source. The radon was inserted from the collecting flask into the exposure chamber by water displacement.

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4.3. Method of Inserting Rats into Chamber.

An apparatus was built for inserting a cage containing a rat into the chamber while continuously sealing the chamber from the outside. It is shown in Fig. 7. The hollow lucite piston in Fig. 7 was constructed to fit as closely as possible into the brass barrel. The rat cage was inserted into the piston through a large opening which was cut in the side. This was the main opening for the rat cage to the chamber atmosphere. Other small air holes were also cut in the lucite cylinder. The brass cylinder had two outlets through which room air could be flushed to expel the radon in the cylinder out-of-doors.

4.4. Sampling for Radon Daughter Products.

Air from the chamber was taken from an opening close to and on the same level with the rat cage. The hole was closed by a metal slide which was clamped between two felt gaskets. A rubber stopper could be fitted into a lip outside the slide. Glass tubing and other equipment was connected to the chamber through this stopper. The daughter products in the chamber air were removed with an AA millipore filter. Two sampling methods were used.

Method 1. Grab Sampling. A liter bulb was evacuated and connected to the outlet side of the millipore holder whose inlet side was connected to the chamber. On opening the flask, air was sucked into the bulb through the millipore, which filtered out the

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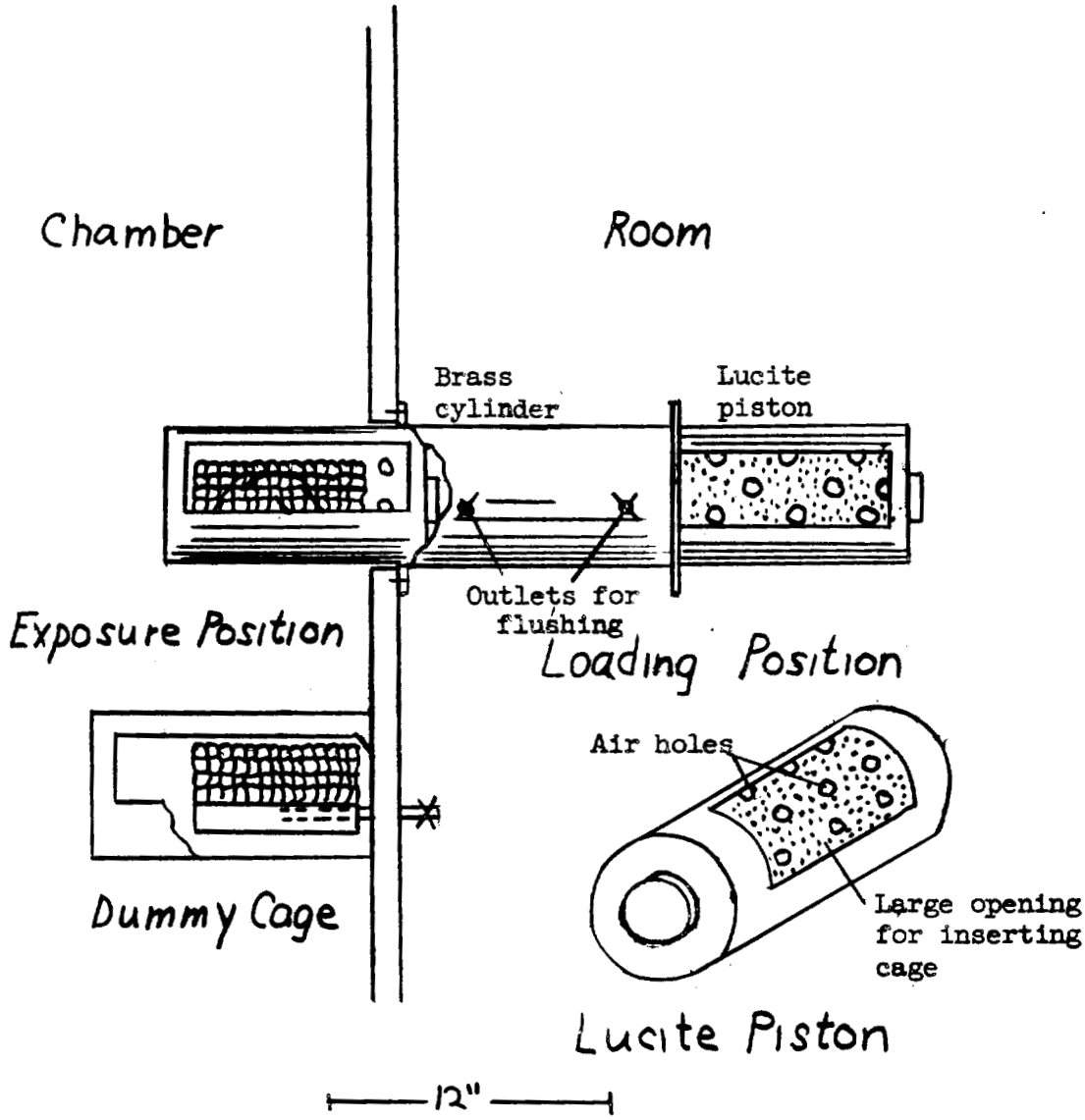

Fig. 7. Method of exposing rat in chamber.

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decay products. In most of the runs a capillary was inserted between the bulb and the filter paper to allow for more uniform sampling. A liter was collected in about fifteen seconds through the capillary.

An alternate method was used at times for grab sampling. A large jar filled with water was placed at some distance above the sampling outlet and the top connected to the millipore holder. A rubber tube from the bottom of the jar passed down several feet to a liter graduate. One could pass a liter of air through the millipore in about fifteen seconds by using this water displacement aspirator and letting the water fall freely through the tube into the graduate. An extra half minute was allowed after the graduate was filled with water to insure equilibration of pressures on both sides of the millipore filter.

Method 2. Slow Sampling. Slow sampling was accomplished by attaching a capillary to the outlet tube of the water aspirator, causing the water to flow out very slowly. The sampling rate was approximately 0.1 liters per minute.

The sampling apparatus is shown schematically in Fig. 8.

4.5. Source of Inhaled Air in Dog and Human Experiments.

In both the dog and human experiments air was breathed from the sampling outlet described in Section 4.4. Detailed descriptions of the experimental arrangements are given in Sections 8 and 9.

4.6. Measurement of Radiations.

Alpha radiation from the millipore filter was measured with a scintillation counter. The scintillation counter was calibrated with an uranium foil whose absolute counting value was determined by

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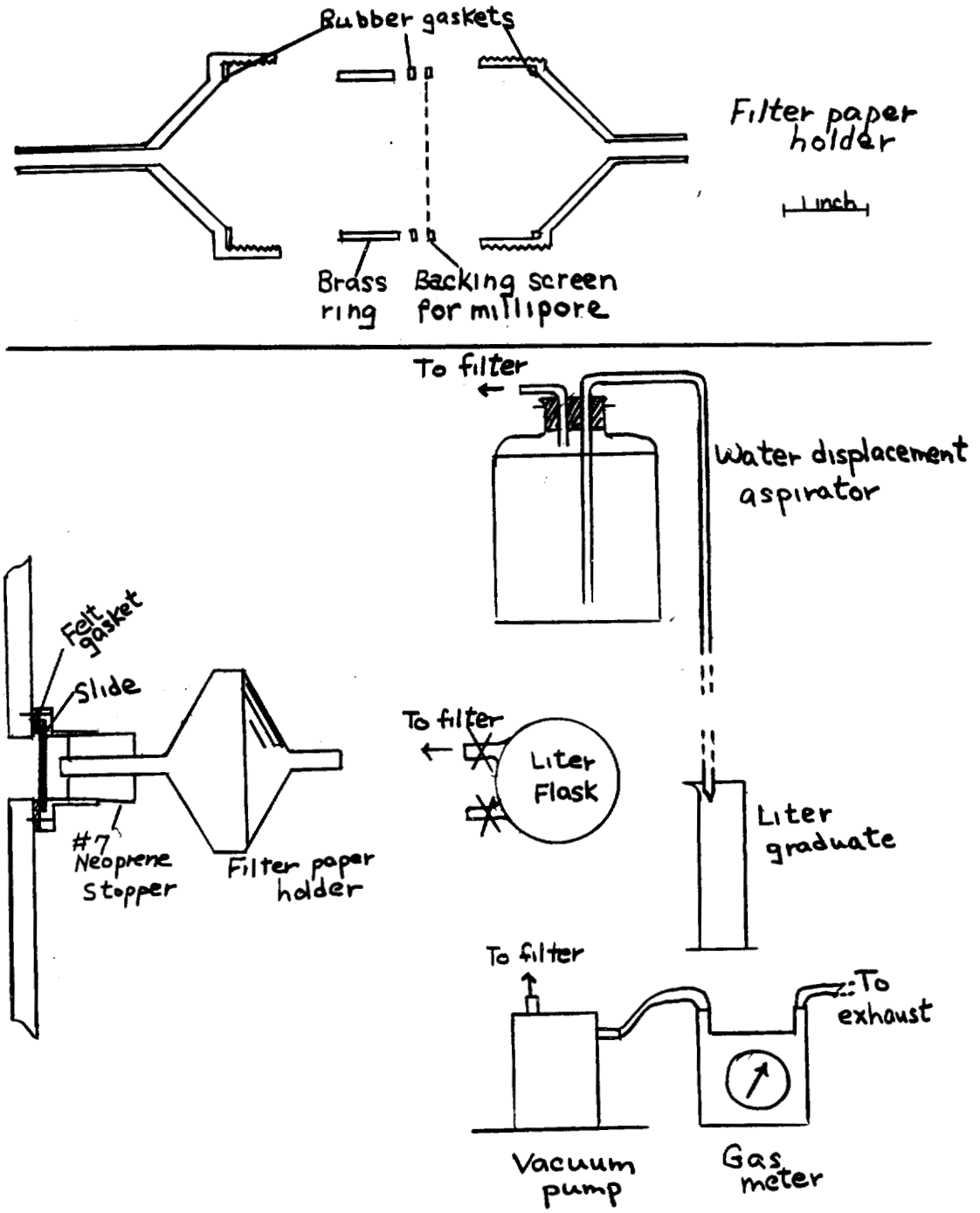

Fig. 8. Air sampling methods.

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measurement in a 2π gas flow proportional counter. The efficiency of the scintillation counter was about 42 per cent.

Radon in the chamber was measured with the vibrating reed electrometer. The radon determinations were made on the same air samples that had been drawn through the millipore filter for daughter product analysis.

The radon from the liter bulb was shared with a 1.8 liter ionization chamber which was connected to a vibrating reed electrometer. It did not appear necessary to add any air to the chamber to bring it up to atmospheric pressure. Operating at a lower pressure decreased the sensitivity of the apparatus somewhat. This was unimportant because the measured radon levels were high. The method gave good reproducibility.

In the early experiments, a cold trap was inserted between the liter bulb and the millipore filter to dry the gas before inserting it into the ionization chamber used with the vibrating reed electrometer. This is standard procedure in radon measurements with the alpha counting apparatus used for low level radon determinations at the University of Rochester Atomic Energy Project. The counters respond to electrons and it is not desirable for the electrons to become attached to the water vapor molecules as this slows the collection time. Since the vibrating reed was operated as a current reading instrument rather than as a pulse counter, there did not appear to be any reason why the air should be

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completely dried, and drying of the air was eliminated in later runs. The later runs did not yield any noticeably different results from the previous measurements where the air had been dried.

The radon measuring system was calibrated by deemanating a two microcurie radium sample provided by Dr. John B. Hursh. The radon from the standard source was collected in the liter bulb and shared with the ionization chamber of the electrometer in the same manner as radon samples from the exposure chamber. The results of two calibration tests were:

Buildup time of radon	Decay time after collection	mv	Curies/mv
3 hr.,42 min.	8 hr.	245 ± 3	21.5×10^{-11}
12 hr.	3 hr.	785 ± 3	21.56×10^{-11}

The calibration figure of 21.5×10^{-11} curies per mv. was used for this method of determining the radon.

Gamma measurements on the rats' lungs were made with a well-type scintillation counter using a sodium-iodide crystal after the design of Anger (30). The first counter used was calibrated with I^{131} and Fe^{59} solutions provided by the Radioisotope Services Division of the University of Rochester Atomic Energy Project. The strength of the I^{131} solution had been determined by the Radioisotopes Services Division. The Oak Ridge value for the radioactivity of the Fe^{59} was used. It was believed accurate to within 10 per cent.

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The results of efficiency determinations as a function of sample volume for both of the isotopes are given in Table 4. The gamma activity of a uranium ore source, made by sealing some uranium ore powder into a brass rod, was noted at the time of the efficiency determinations and the source was subsequently used as a standard to calibrate the counter from day to day. The gamma spectra emitted by the daughter products and the standard were similar enough to make this calibration method suitable for the purposes of the present investigation.

The efficiency of the counter used on the first experiments was approximately 10 per cent. An improved counter used on the later experiments had about double this efficiency. It was calibrated by a method described in Section 7.3.

Table 4.

Efficiency of NaI Well-type Scintillation Counter for I^{131} and Fe^{59} Solutions as a Function of Sample Volume.

Volume in Test Tube cc.	Height in Test Tube mm.	Per cent Efficiency	
		I^{131}	Fe^{59}
0.5	8	13.6	12.3
1.0	15	12.6	11.8
1.5	22	11.3	9.7
2.0		9.7	8.9
2.5		8.9	
3.0		7.9	
3.5		7.1	
4.0		6.1	
5.0		5.2	

Uranium ore standard = 4270 cpm

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A constant voltage transformer was used to improve the voltage stability of the crystal counter since the counting rate was very senitive to fluctuations in the high voltage supply.

The crystal was shielded by about three inches of lead. This was very effective for shielding gamma radiation. However, at the first location of the counter, the background rose by a factor of ten when the 130 in. cyclotron at the University was on. This was possibly due to n, γ reactions in the lead shield. A location was found in the building at which the background rose only 50 per cent when the cyclotron was on. When the cyclotron was off, the background was 120 cpm. When it was desirable for the background to remain as constant as possible, the experiments were performed on those days when the cyclotron was off.

The Texaco well-type Geiger counter, Type GR, manufactured by Welch Allyn Inc., was used for the experiments on the dog. It had a background of 500 cpm.

5. SAMPLING TESTS WITH MILLIPORE FILTERS.

Millipore filter paper, type AA, was selected for the sampling. Its properties have been described by First (31). The AA filter has an estimated pore size between 0.5 and 0.7 mu. According to First, it may be used as a sieve under gentle suction, retaining particles greater than 0.9 mu. When air is pulled through at a rapid rate, however, the filter is effective down to

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extremely small particle sizes. This is explained as due to an electrostatic action produced when the air rushes past the filter surface.

The millipore filter becomes transparent when immersed in cedarwood or special oils which have the millipore index of refraction of 1.56 and thus it can be examined under the microscope. The millipore sample is prepared for examination by placing it, dust side down, on a clean microscope slide and adding a few drops of oil to the upper side. Special techniques have been described for examining millipore filters under the electron microscope (31, 32).

Tests were conducted on the millipore sampler, using the grab and the slow sampling methods described in Section 4.3. Both methods were considered useful for measuring the daughter product concentration in the chamber air. The grab sample contained the material filtered from air which had been sucked through the filter paper very rapidly into an evacuated liter bulb. It therefore gave the filterable decay product activity per unit volume of chamber air. The slow sample was obtained by drawing the air at approximately the same rate at which the rats were breathing (i.e. 100 cc/min). The slow sample, therefore, might give the actual daughter product concentration breathed by the experimental animals more accurately than the grab sample. Accordingly, experiments were conducted at various times during the course of the

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investigation to answer the following questions:

1. Did the millipore filter give reproducible results for repeated measurements on the same atmosphere?

2. How efficient was the filter paper in removing the decay products from the atmosphere? What was the loss in alpha counts because of self-absorption in the filter paper?

3. Was there any loss of decay products in the sampling tubes preceding the millipore filter?

4. Did the decay product concentration in the air fluctuate to any extent about a mean value over short periods of time? If this occurred, one should prefer to sample over a relatively long period than to take a grab sample.

5. How did measurement of the decay product concentration differ when the air was drawn through the millipore at a fast rate as compared to when it was drawn through slowly?

The results of the tests depended on the dust load in the air that was sampled. The most consistent and reproducible results were obtained when the radon was mixed with air which had sufficient dust in it so that radioactive equilibrium or near equilibrium conditions existed among the radon daughters in the chamber. Such air was obtained by placing a fan in an open window which was next to the chamber, and blowing air into the chamber directly from the outside. The laboratory building was located near the university powerhouse and the animal house incinerator, and since the odor of

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smoke was almost always in the air, it may be assumed that these two sources kept the air well supplied with nuclei on which the radon daughters could be deposited.

5.1. Tests of Grab Sampling Technique -- Radon Mixed with Air Taken Directly from the Out-of-Doors.

1. Reproducibility. Two grab samples taken in succession always agreed very closely, and the differences were no greater than the differences to be expected from the statistical variations in the counts. The results of one group of experiments are given in Table 5. The samples were taken four minutes apart. In runs 1 and 3, the air flow through the filter paper was slowed by inserting a capillary between the filter and the evacuated bulb. The bulb was filled with air about fifteen seconds after it had been opened to the chamber. In runs 2 and 4, no capillary was used and the bulb was filled very quickly. Forty seconds were allowed for equilibration between the bulb and the chamber in all four runs. The activities obtained when the capillary was used were the same as when the capillary was not used.

2. Efficiency of sampling. An idea of the efficiency of filtration was obtained by putting two filters in series and comparing the activity picked up on the second filter to the activity picked up on the first. When the series filters were about 3/16 in. apart, negligible activity was measured on the second filter paper.

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Table 5.

Reproducibility of Measurements of Daughter Product Activity with Grab Sampling Method

Run	Method[a]	Counts
1	Cap	7477
2	No cap	7536
3	Cap	7281
4	No cap	7189
	Runs 1 and 3 Avg.	7379
	Runs 2 and 4 Avg.	7363

[a]Cap. Sample drawn through with evacuated liter bulb, with capillary between bulb and filter to give more uniform sampling rate.

No cap. Sample drawn through with evacuated liter bulb.

In one run, the readings of the first and second filters were 7600 and 5, respectively.

The loss of counts from the filtered daughter products because of self-absorption in the millipore filter was obtained by taking three measurements on the millipore sample. Measurement 1 was the normal measurement of the sample activity. Measurement 2 was the activity measured after the sample had been covered with an unused millipore filter. Measurement 3 was the activity measured when the radioactive millipore filter was placed in the counter upside down. The equation relating these three readings and the ratio of measured activity to true activity is derived

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in Appendix 1. It is

$$\frac{\text{Measured activity}}{\text{True activity}} = \frac{2 \times (\text{Meas. } 1)}{2 \times (\text{Meas. } 1) + (\text{Meas. } 3) - (\text{Meas. } 2)}$$

The results of the self-absorption measurements are given in Table 6. The average loss of counts from self-absorption was 3 per cent.

Table 6

Measurement of Self-Absorption Loss in Millipore Filter Paper Sample

Run	Meas. 1	Meas. 2	Meas. 3	Measured Activity / True Activity
1	8690	5478	5710	0.985
2	6753	3484	4130	0.955
3	15250	9800	10750	0.97

Meas. 1. Direct measurement.
Meas. 2. Activity after clean millipore placed on top of sample.
Meas. 3. Activity of sample turned upside down.

3. Losses in sampling lines. Studies were made only under slow sampling conditions. Because of the small size of the nuclei on which the daughter products were carried, they were removed from the air stream mainly by diffusion to the walls and by sedimentation to the bottom of the sampling tube. Therefore the probability of contact of a particle with the wall of the tube increased with the time spent by the particle in the tube. It will be shown in a later paragraph that even under slow

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sampling, the losses in the sampling lines used were negligible. It therefore followed that losses in the sampling tubes were also negligible under grab sampling conditions.

4. Variations of activities of samples taken at short time intervals. Table 7 presents the results of repeating measurements of the daughter product activity in the chamber at different time intervals. The table shows that the daughter product concentration changed only slightly over a period of several hours.

5. Effect of velocity of air flow on filtering efficiency. Sampling the chamber air by the evacuated bulb method or by the use of the water displacement aspirator gave the same results when the liter was drawn through in less than fifteen seconds. In one set of experiments on air to which an aerosol had been added to raise the air-borne daughter product activity, the activity removed from a liter of air drawn through the filter paper by the water displacement aspirator in 1.5 minutes, was 94 per cent of the activity removed from a liter drawn through with the evacuated bulb.

5.2. Tests of Slow Velocity Sampling Technique.

Two filter papers were compared at a slow sampling speed for reproducibility of measurements on air containing normal atmospheric dust by connecting the common end of a Y-tube to the chamber and connecting the ends of the Y to the two filters. The outlets of the filter holders were connected to two water

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Table 7.

Variations in Daughter Product Activity in Exposure Chamber Over Short Time Intervals

Date	Exp. Cond.[a]	Time sample taken(min.)	Counts/min.
4/21	I-2	0	6100
		30	6700
		120	5400
5/7	I-18	0	3600
		15	3450
7/27	I-1	0	17800
		60	17300
		270	17800
7/31	I-5	0	5900
		60	5840
		78	5750
		96	5550
		110	5500
8/3	I-8	0	4300
		3	4200
		6	4300

[a]See Table 9.

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displacement aspirators. Thus, both millipores sampled from the same tube. The sampling rate was 0.1 liters per minute. The results of one set of measurements are given in runs 2 and 3 of Table 8. The activities agreed to one per cent. They were 9 per cent lower than the activity of sample 1, which was the same size as samples 2 and 3 but taken sixteen minutes earlier.

Table 8.

Reproducibility of Measurements of Daughter Product Activity in Exposure Chamber for Slow Sampling Rate (0.1 l/m)

Run	Sampling time (min.)	Activity
1	0-10	7650
2[a]	16-26	6910
3[a]	16-26	7080

[a]Runs (2) and (3) were simultaneous samples of the same air.

No significant difference was measured between the activity removed from air drawn directly from the chamber and air which first passed through a 5/16 in. diameter, 10 in. long tube at the 100 cc. per minute sampling rate. In another experiment, air was drawn through a 15 in. long, 0.042 in. I. D. Genflex plastic tube (#603), placed immediately preceding the millipore filter. The flow rate was 100 cc. per minute. The tubing was then coiled up and counted in the same gamma counter as the filter paper. The activity deposited in the tube was about one per cent of the

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activity removed by the filter paper. The experiment was conducted two days after air had been inserted into the chamber from the out-of-doors.

Since the decay of a radioactive sample is linear over time intervals which are short compared to the half-life, the activity of a liter sample taken over ten minutes should very nearly equal the activity of a liter sample taken over a few seconds if the time of decay of the ten minute sample is measured from the middle of the sampling period. In practice, the activity of the ten minute sample was always less than the activity of the grab sample, and the difference was greater the lower the dust load in the chamber. The ratios of the activities of slow to grab samples as measured at various times during the course of the investigation are given in Table 9.

All the values given in Table 9 are for samples taken from an opening in the chamber wall. Three factors were studied for their relative importance in affecting the differences in activities obtained for slow and grab sampling.

1. The slow sampling method drew air from a region that was closer to the wall and thus perhaps had a significantly lower daughter product concentration than the region sampled by the grab sample.

2. The difference was because of poorer filtration at the slower velocities through the millipore filter.

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Table 9.

Ratios of Activities of Slow to Grab Air Samples

Date	Exp. Cond.[a]	Slow/Grab
4/20	I-1	0.88
4/21	I-2	0.83
4/22	I-3	0.42
4/23	I-4	0.29
7/27	I-1	0.85
7/28	I-2	1.02
8/1	I-6	0.83
11/12	I-1	0.49
11/13	I-2	0.39
	Fan on 10 min.	0.64
	Fan on several hours	0.78
	Fan off	0.67
	Fan on 10 min.	0.78
11/14		0.83
11/17	Fan on all previous night	0.84
11/23	II-1	0.07

[a]Experimental Conditions

I Outside air blown into chamber during previous night and early morning of first day of series of experiments. Chamber then sealed and radon inserted. No changes for succeeding experiments. Air not stirred. Arabic numeral gives day of experiment counting from time air changed in chamber.

II Filtered air blown into chamber during night preceding experiment.

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3. The difference was because of increased deposition on the walls of the filter holder, since there was a dead space of about 30 cc. in the filter holder itself. At a flow rate of 100 cc. per minute the air would have remained approximately twenty seconds in the small volume of the filter holder before passing through the filter.

The wall effect was eliminated as a major cause of the difference in the slow/grab ratio by comparing the ratio for air sampled directly from the hole in the chamber and for air sampled from a tube leading to a point about twelve inches inside the chamber. The experiment was done when filtered air was in the chamber, a condition which gave the greatest difference between slow and grab sampling. There was no significant difference in the slow/grab ratio for samples taken from the region next to the wall and for samples taken from the region twelve inches inside the chamber, although the measured activity at the wall was only about 59 per cent of the measured activity inside the chamber.

The effect of linear flow velocity through the millipore filter on its filtering efficiency was studied in two ways. In the first method, the activities of two millipores which had been placed in series about 1/8 inch apart were compared. The experiments were done in a filtered air atmosphere. For

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fast sampling, no activity was found on the second millipore paper. For slow sampling, the activity of the second filter was about 7 per cent the activity of the first filter.

Another method used was to sandwich the millipore filter between two rubber diaphragms, one centimeter in diameter, to increase the linear flow velocity through the millipore. Experiments were done in a filtered atmosphere. Since the utilized diameter of the millipore when used in the filter holder without the diaphragm was 3.7 cm., the flow velocity through the smaller opening was fourteen times as great as through the larger. The difference in activities was compared for a 1.5 minute liter sample through the larger area, and a ten minute liter sample through the smaller opening. The ten minute sample activity was 18 per cent of the activity of the 1.5 minute sample, which in turn was 51 per cent of a grab sample. In the other experiments where a diaphragm was used with ten minute liter samples to increase the flow velocity through the millipore, the activity was always lower than when no diaphragm was used for the same sampling minute volume. These results indicate that the difference in linear velocity through the filter was not the major factor in producing the difference between the slow and grab sampling.

The only remaining factor that could account for the difference between slow and grab sampling was increased deposition

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on the filter holder at the slow rates. In one experiment aluminum foil was used to line the millipore holder and air was sampled through at the slow rate. The activity measured on the foil at the end of the sample period was not enough to account for the difference between slow and grab sampling. However, the measurements were very rough and it is possible that the value of the deposition in all regions of the filter holder when measured more accurately will account for a large share of the difference between the slow and the grab samples.

5.3. Conclusions from Sampling Tests.

Because the activity of a slow filter paper sample of a given volume of air was always less than the activity of a grab sample, and because the difference increased as the dust load decreased, the slow sample was not a reliable method of determining the daughter product concentration in the chamber air. All the values for the RaC' concentrations in the chamber were obtained from grab samples using the sampling outlet described in Section 4.4.

One explanation for the dependence of the difference between slow and fast sampling on the dust load in the chamber is that under low dust conditions the daughter products were suspended in the air in a very mobile form and were removed to a considerable degree by the walls of the filter holder when the air was drawn

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through slowly. Since the mobility of the daughter products plays an important role in determining their deposition in the respiratory tract, the ideal sampling method would give their mobility as well as their concentration in the sampled air. Although it seems that the slow sampling method could be developed to give an indication of the mobility of the suspended daughter products, it was not studied sufficiently in the present investigation to present as a useful supplement to the grab sampling method.

6. OBSERVATIONS ON THE DEPARTURE OF THE FILTERABLE DAUGHTER PRODUCTS FROM EQUILIBIRUM WITH THE RADON IN THE CHAMBER AIR.

6.1. Departuresfrom Equilibrium in Stored Atmospheric Air and in Filtered Air.

An analysis of the decay curve of the daughter products removed from an air sample taken from the chamber several hours after the insertion of outdoor air usually showed radioactive equilibrium between RaA, RaB, and RaC-C'. Decay curves of samples taken over a period of days after the radon was inserted showed a gradual steepening in the first part of the curve, indicating an increased proportion of RaA, and a gradual leveling off of the central portion of the curve, indicating that RaC-C' was decreasing with respect to RaB. This can be explained in terms of the decrease of the atmospheric dust in the chamber air over this time. As the dust concentration decreases, the walls compete to a larger extent for the radon

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daughter products because their surface area increases relative to the surface area of the dust. Because the later elements in the decay series have a greater chance to get to the walls than the earlier elements, their concentrations will decrease relative to the earlier ones as the dust concentration decreases. Decay curves taken over a period of days are given in Fig. 9 along with the calculated fraction of equilibrium with radon attained by RaA, RaB and RaC-C'.

Table 10 summarizes the departures of the daughter products from equilibrium with the radon in the chamber air, expressed as the per cent of the total RaC' in the chamber which was filterable from the chamber air. Series I, III, and IV give three different sets of experiments in which the RaC' concentration was measured on successive days starting from the day outdoor air was put into the chamber. Series II gives the results of sampling from the chamber after it had been filled with filtered air. The concentration averaged 5.6 per cent of the concentration which would be in equilibrium with the radon as compared to a high of 81 per cent measured in freshly inserted uncleaned outdoor air. The lowest RaC' concentration in uncleaned air was measured after the dust had settled over a period of two weeks. The RaC' dropped to 10 per cent. Stirring the air brought it down to 1.6 per cent.

All the values in Table 10 were obtained by extrapolating the measured alpha activity on the filter paper to zero time, assuming equilibrium between RaA, RaB and RaC-C' in the chamber. Equilibrium often did not exist and the error involved in neglecting the true

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Fig. 9. Measured decay of liter grab samples of radon daughter products as a function of settling time of dust in chamber. Outside air blown into chamber during previous night and early morning of first day of experiments (4/20). Chamber then sealed and radon inserted. All data extrapolated to same radon level of 10000 dpm.

The vertical lines through the experimental points give the 95% confidence intervals

The curves presented in conjunction with each series of decay measurements is a theoretical curve which closely fits the measured data and which was calculated for the initial relative concentrations of RaA, RaB, and RaC-C' given by the first, second, and third digits on the curve.

The experimental data on the curve dated 11/24 is from an independent experiment in which the chamber was filled with filtered air before the radon was inserted.

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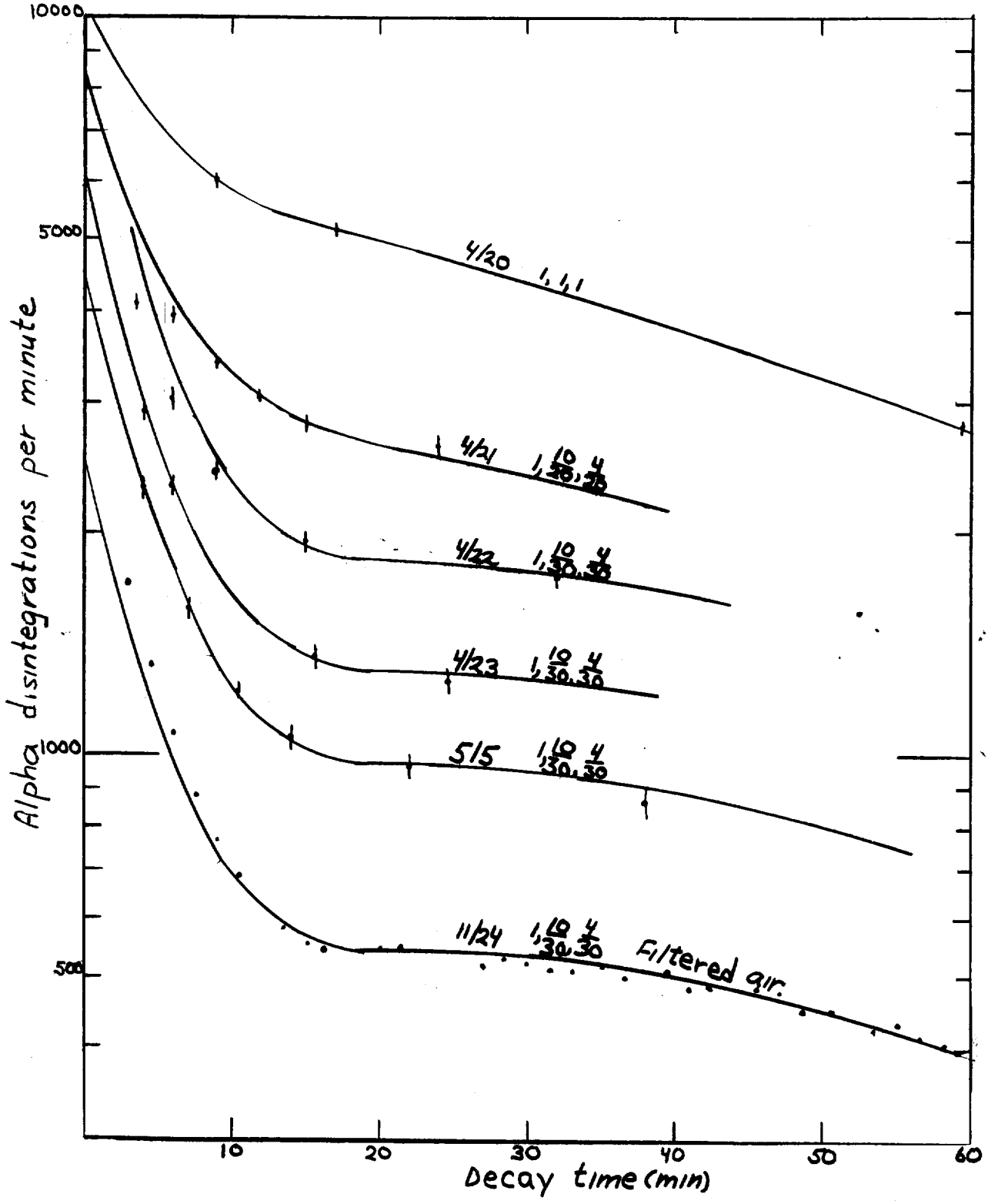

Fig. 9

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Table 10.

Approach to Equilibrium with Radon Attained by RaC' in Air of Exposure Chamber

Days air in chamber	Series I % Equil.	Series II % Equil. (air filtered)	Series III % Equil.	Series IV % Equil.
1	52	5	61	81
	41	6.8		
2	42	air fanned 2.2	34	77
	39	1.6		
3	26	2.2	25	
	27			
4			16	
6				36
			ZnO_2 dust added	79
8				46
			Air fanned	4
			Fan turned off	32
13			13.2	
		Animal experiment done		
14			20	
15			23	
		Cigarette smoke added		
			94	

RaC' measured was removed from air with millipore filter; Rn was measured with vibrating reed electrometer.

Series numbers refer to chronological order of experiments.

In Series I, III and IV the measurements were taken on successive days after outside air was blown into chamber; in series II, the air in the chamber was replaced with filtered air.

Beginning dates. I. 10/31; II. 12/13; III. 4/20; IV. 7/27.

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relative concentrations of the daughter products will be discussed in Section 6.4. It will be shown that from the standpoint of calculating the dosage to the lung produced by breathing the daughter products over an extended period of time, the error involved in neglecting the relative departures of the daughter products from equilibrium is small.

6.2. Departure from Equilibrium in Fanned Air.

In one experiment, atmospheric air which had been stored in the chamber for several days was fanned and sampled periodically. The fan was on the floor in one corner and pointed toward the opposite corner about half way up from the floor. Decay curves of the activity removed from air sampled during the fanning are given in Fig. 10.

The daughter product acitivity in the chamber decreased by an order of magnitude two hours after the fan had been turned on. The motion of the air thus increased the effectiveness of the walls in removing the daughter products. Two hours after the fan was turned off the activity of an air sample had risen to 70 per cent of the value measured at the beginning of the experiment.

6.3. Departures from Equilibrium in Different Regions of Exposure Chamber.

Because all solid material in the exposure chamber was a sink for the daughter products, it was to be expected that the

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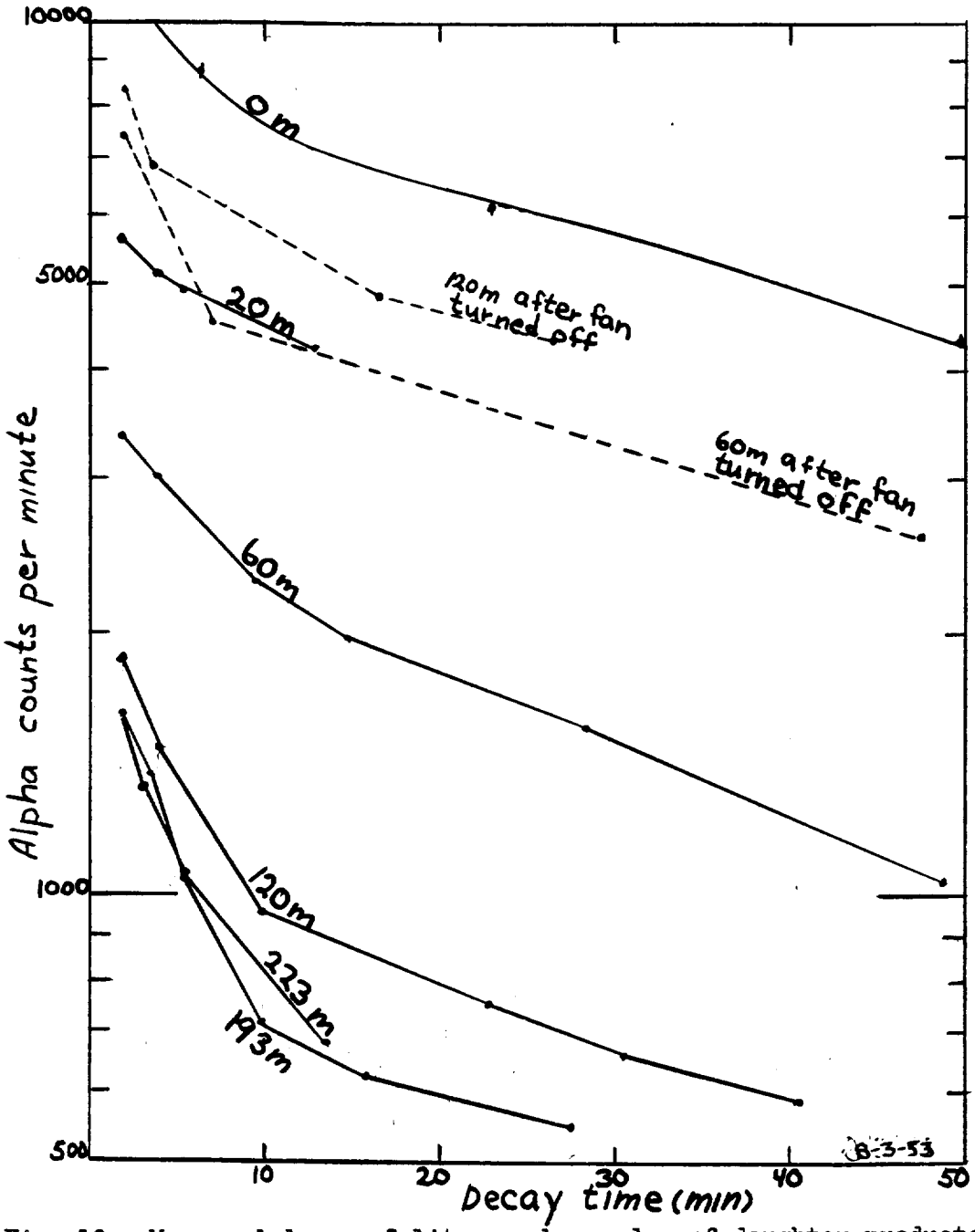

Fig. 10. Measured decay of liter grab samples of daughter products as a function of time air stirred in chamber. Fan turned on at t=0.

The curve presented in conjunction with the measurements on the sample taken before the fan was turned on is a theoretical decay curve for initial radioactive equilibrium between RaA, RaB, and RaC-C'.

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daughter product concentration in the air in any particular region of the chamber would depend on the geometry of the solid matter in the environment. In perfectly still air the solid environment which would be most important in influencing the air-borne daughter product concentration in the region of interest would be that within a distance of the order of magnitude of the distance traveled by the daughter products during their average lives. This is a few centimeters for daughter products which undergo Brownian movement in the air as molecules or as constituents of molecular aggregates, but the distance becomes decreased by several orders of magnitude when they become deposited on the suspended atmospheric dust particles. However, the air is never perfectly still, and its motion serves to extend the sphere of influence of any solid matter in removing daughter products from the air.

Perhaps the most difficult problem in the entire investigation was that of determining the daughter product concentration breathed by the rats. Since the rats were inserted into the chamber in wire cages, which in turn were partially enclosed in a lucite cylinder, they breathed in an environment which under certain conditions might contain a daughter product concentration much lower than that existing in more open regions of the exposure chamber. The results of sampling experiments from the different regions in the exposure chamber are given in Table 11. Region 1 was air taken directly from an opening in the wall of the chamber.

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Table 11.

Relative Air-borne Daughter Product Activities in Different Regions of Exposure Chamber

Date	Exp. Cond.[a]	Slow Grab[b]	Region from Which Air Sampled: I Hole in wall	II Tube 12" into chamber	III Dummy cage in chamber
9/2	I-1	~0.8	1		0.89
11/23	II-1	0.07	1	1.7	
11/24	II-2	0.16	1	1.6	0.2

[a]See Table 8

[b]Ratio of daughter product activity removed from liter sample collected in ten minutes to activity from liter sample collected in several seconds.

Region 2 was air taken through a tube extending about twelve inches into the chamber. Region 3 was from a dummy cage placed in the chamber in a cylinder in an arrangement similar to that used in the rat experiments (Fig. 7).

It is seen that under normal atmospheric dust loads, the cage activity was about 90 per cent of the daughter product activity in the chamber. However, when filtered air was inserted in the chamber, the activity removed from air sampled from the cage was only 20 per cent of the activity of air removed from the exposure chamber.

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6.4. Errors in Interpreting Sampling Results When Relative Concentrations of Daughter Products Are Not Taken into Account.

The daughter product concentrations in the chamber were determined by measuring the alpha disintegration rate of the filter paper at a known time after the air sample had been taken and extrapolating the measurement back to zero time on the assumption that RaA, RaB, and RaC-C' were in radioactive equilibrium. These concentrations were then used to calculate the daughter product buildup on a filter paper which sampled the atmosphere for an infinite length of time at a specified sampling rate. The results of this calculation could be used to obtain the daughter product buildup and alpha energy released in any filtering systems (i.e. the lung) whose sampling rate and filtration efficiency were specified.

In practice the daughter products were not always in equilibrium. It will be shown, however, that the error in assuming equilibrium to calculate the alpha disintegration rate of an infinite sample is small for the relative air-borne daughter product concentrations measured in practice.

A sample containing equilibrium amounts of RaA, RaB, and RaC-C', each initially giving 10 dpm will give a total of 9.26, 7.10 and 5.05 alpha dpm in 20, 40, and 60 minutes respectively. These alpha dpm would also be measured for the non-equilibrium initial disintegration rates listed in Table 12. The rates given in Table 12

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Table 12.

Various Combinations of Daughter Product Activities Producing Same Alpha Activity After Given Decay Time

I Alpha dpm measured after listed decay time		II Initial combinations of daughter product dpm which will produce alpha dpm in I		
		RaA	RaB	RaC
9.26 dpm 20 min. after sample collected	a)	10	10	10
	b)	23.8	11.9	7.2
	c)	38.3	12.8	5.1
	d)	191	0	0
7.10 dpm 40 min. after sample collected	a)	10	10	10
	b)	21.4	10.7	6.42
	c)	32.2	10.7	4.29
	d)	148	0	0
5.05 dpm 60 min. after sample collected	a)	10	10	10
	b)	20.3	10.2	6.1
	c)	29.5	9.83	3.93
	d)	122.8	0	0

apply to the grab samples of atmospheres containing the following relative concentrations of RaA, RaB, and RaC-C'.

	RaA	RaB	RaC-C'
a)	1	1	1
b)	2	1	0.6
c)	3	1	0.4
d)	1	0	0

Case (d) is an extreme case which would never be encountered in practice.

Table 12 was calculated as follows. Using the above sets of numbers to fix the relative non-equilibrium initial disintegration

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rates of RaA, RaB, and RaC-C', arbitrary values for the magnitudes of the initial disintegration rates were assigned. Thus, for the ratios 3/1/0.4, it was convenient to assign the initial disintegration rates of 30, 10, and 4 to RaA, RaB, and RaC-C' respectively. Fig. 2 was then used to determine the alpha disintegration rate at the desired decay time for the specified initial disintegration rate. Thus 30 dpm of RaA at t=0 gives 0.3 dpm of RaA and 0.38 x 3 or 1.14 dpm of RaC' at t=20. Similarly 10 dpm of RaB give 3.84 dpm of RaC' and 4 dpm of RaC give 0.4 x 4.96 or 1.98 dpm of RaC' at t=20. The total alpha rate is 7.26 dpm. Since 10 dpm each of RaA, RaB, and RaC will give 9.26 dpm at t=20 (Fig. 2 or Fig. 4), the initial non-equilibrium rates which will give the same values are 9.26/7.26 times the arbitrarily chosen values. Thus 38.3, 12.8, and 5.1 dpm of RaA, RaB and RaC-C' will also give the desired counting rate at t=20.

The first set of equations of Section 3.2 can be combined to give a single equation for the buildup of the alpha activity on infinite filter paper samples of atmospheres containing given daughter product concentrations. The equation is

$$\text{alpha dpm} = (2Q_2 + Q_3 + Q_4)\, v$$

Q_2, Q_3, Q_4 = atoms of RaA, RaB, RaC per liter, respectively.
v = sampling rate in liters per minute.

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This equation can also be written

$$\text{alpha dpm} = \left(\frac{2\lambda_2 Q_2}{.227} + \frac{\lambda_3 Q_3}{.0259} + \frac{\lambda_4 Q_4}{.0352} \right) v$$

$\lambda_2 Q_2 (\lambda_3 Q_3, \lambda_4 Q_4)$ = RaA (RaB, RaC-C') dpm per liter
v = sampling rate in liters per minute.

Table 13 gives the buildup after an infinite sampling time on a filter paper which removes the daughter products with 100 per cent efficiency from the atmospheres containing the concentrations given in Table 12 at a sampling rate of one liter per minute. The table shows that the buildup of the activity on an infinite filter paper sample is closely proportional to the activity of a grab sample, counted preferably one hour after decay, and is very insensitive to the actual relative concentrations of RaA, RaB, and RaC-C' except when the departure from equilibrium is extreme.

Table 13.

Activities of Infinite Samples Calculated from Activity of Grab Sample for Different Non-Equilibrium Daughter Product Concentrations in Atmosphere

Relative concentrations of daughter products in sampled atmosphere			Activities of infinite samples (1 l/min sampling rate) when grab samples give same activity after fixed decay time		
RaA	RaB	RaC	9.26 dpm at 20m	7.10 dpm at 40m	5.05 dpm at 60m
1	1	1	758	758	758
2	1	0.6	873	785	745
3	1	0.4	975	821	752
1	0	0	1680	1305	1080

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For a specific example, suppose a liter grab sample of the daughter products is taken and after it has been allowed to decay for 60 minutes, gives 5.05 dpm. If we assume the daughter products are in equilibrium in the sampled atmosphere, we see from Table 12 that the activities of RaA, RaB and RaC-C' in the sampled atmosphere are each 10 dpm per liter. We read from Table 13 that for air sampled at the rate of one liter per minute an infinite sample would build up to an alpha distintegration rate of 758 dpm.

We will now show that very similar results are obtained in sampling atmospheres not in equilibrium. The greatest departures from equilibrium measured in American uranium mines by Holaday et al (25) were not as great as the ratios 3/1/0.4 for RaA/RaB/RaC-C' used for the calculations in Tables12 and 13. For the 3/1/0.4 relative concentrations, the extrapolation of the 5.05 dpm sample to zero time gives 29.5, 9.83, and 3.93 dpm per liter for RaA, RaB and RaC-C' respectively. The calculated buildup of an infinite sample of this atmosphere at a one liter per minute sampling rate is 752 dpm. The difference between this result and the result assuming equilibrium is less than one per cent. On the other hand, it is seen from the table that extrapolations from single readings of grab samples counted after 40 minutes decay can be about 8 per cent low and after 20 minutes decay can be 22 per cent low for the extreme departures from equilibrium which may be encountered in practice.

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Since the lung acts as a filter of the daughter products, the buildup of alpha activity in the lungs of a person breathing in an atmosphere containing the daughter products is not sensitive to the relative daughter product concentrations in the air but depends only on the activity of a grab sample, measured 40 to 60 minutes after it is taken. The average alpha energies per alpha disintegration emitted by the daughter products built up in an infinite sample are calculated from the data in Table 3 and the equations in Section 3.2 to be 7.56 Mev. and 7.4 Mev. for relative air-borne concentration of RaA / RaB/RaC-C' = 1/1/1 and 3/1/0.4 respectively. Therefore, the error in evaluating the total energy imparted to the lung from a single measurement of a grab sample is also small. The error in evaluating the average energy imparted per gram of lung tissue, which is the important quantity from the standpoint of determining the dosage, is even smaller since the range of the alpha particles in the tissue is approximately proportional to the energy of the alpha particles.

We therefore conclude that the activity of a grab sample of decay products filtered from air and determined at a fixed time 40 to 60 minutes after the sample was taken is an index of the alpha dosage to a human being breathing this air, regardless of the relative concentrations of RaA, RaB, and RaC-C' in the sampled air, if conditions are otherwise equal.

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7. RESULTS OF RAT EXPERIMENTS.

7.1 Procedure.

The arrangement for inserting the rat into the chamber has been described in Section 4.3 and illustrated in Fig. 7. To insert the rat cage into the chamber, the lucite piston was drawn partly out of the brass cylinder to the loading position indicated in Fig. 7 and the cage containing the rat was put in. The piston was then pushed through the cylinder into the chamber as shown in the figure. A lucite cover was screwed on the outside of the brass cylinder.

After a period of about 2-1/2 hours, the piston was drawn into the cylinder and room air flushed through for one minute. This was considered the zero point of the daughter product decay in the lungs of the animal. The piston was then drawn partly out of the cylinder and the cage removed. The animal was anesthetized in ether and the lungs removed. The lungs were then inserted into test tubes and counted in the well-type counter. During the time the animal was exposed, filter samples were taken to determine the daughter product levels in the chamber. The animals were usually resting quietly most of the time they were in the chamber.

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7.2. Variation of Radioactivity of Lungs with Daughter Product Activity in Air.

The experiments were conducted using twenty-two male rats of the Wistar strain over a period of several months. The rats were exposed to radon mixed with the following atmospheres:

1. air inserted into the chamber from the out-of-doors a few hours before the animal exposure;
2. air confined in the chamber for several days before the exposure;
3. filtered air;
4. air to which cigarette smoke had been added;
5. air to which a fine ZnO_2 dust had been added.

During most of the experiments, the air in the chamber was undisturbed. In some, a small fan was turned on for several hours preceding and during the time the rat was in the chamber. The results are given in Tables 14 and 15.

The RaC' values in Tables 14 and 15 were calculated from one measurement of a grab air sample taken from an opening in the chamber wall on the assumption that radioactive equilibrium existed between RaA, RaB, and RaC-C'. It was seen from Table 13 that even when the departure of the daughter products from equilibrium is fairly large, the assumption of equilibrium may be used to calculate the activity of an infinite sample. When the air in the chamber

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Table 14.

Results of Rat Experiments. Counter I

Run	Date	Exp. Cond.[a]	Rat Wt. gm.	Rn Conc. $c/\ell \times 10^9$	Activity of lungs γ dpm[b]	RaC' Conc. $c/\ell \times 10^9$	Lung Activity / RaC'
1	11/4	I-1	189	24.1	20700	12.7	1630
2		I-1	233	23.4	66600	10.9	6110
3	11/5	I-2	197	18.8	20600	8.07	2560
4		I-2	210	18.4	15200	7.32	2080
5	11/6	I-3	190	15.5	13050	4.08	3200
6		I-3	177	14.7	11300	4.06	2780
7	12/13	II-1	175	25.2	3560	1.22	2920
8		II-1	198	24.9	3780	1.69	2240
9		II-1	170	24.9	3330	1.27	2620
10	12/14	II-2s	184	20.2	490	0.272	1800
11		II-2s	189	19.8	312	0.349	895
12	12/15	II-3s	180	16.3	342	0.116	2940

[a]Experimental Conditions

I Outside air blown into chamber during previous night and early morning of first day of series of experiments. Chamber then sealed and radon inserted. No changes for succeeding experiments. Air not stirred.

II Chamber washed, lined with glycerine-coated glossy paper. Filtered air blown into chamber during night preceding first day of series. Chamber sealed and radon inserted on first day.

's' means air fanned. Arabic numeral gives day of experiment counting from time air changed in chamber.

[b]Corrected to 100% detection efficiency. Approximately ten times measured counts per minute.

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Table 15.

Result of Rat Experiments. Counter II

Run	Date	Exp. Cond.[a]	Rat Wt. gm.	Rn. Conc. c/ℓ x 10^9	Activity of lungs γ dpm[b]	RaC' Conc. c/ℓ x 10^9	Lung Activity / RaC'
13	4/20	I-1	219	24.3	87500	13.3	6580
14	5/5	I-16	215	22.8	12600	2.68	4700
15		I-16	196	22.2	25600	~7.5	3420
16	5/6	I-17s	189	16.5	420	low	
17	5/8	I-19[c]	197	21.0	53800	19.1	2820
18	7/27	I-1	214	22.4	76500	20.3	3740
19	7/31	I-5	247	22.6	27400	7.61	3600
20	7/31	I-5[d]	230	22.2	72500	16.9	4290
				Results with Restrained Rats			
21	9/2	I-1	195	22	88500	21.2	4170
22		I-1	200	22	88500	21.2	4170

[a]See Table 14.

[b]Corrected to 100% detection efficiency. Approximately five times measured counts per minute.

[c]Cigarette smoke added

[d]ZnO_2 powder added

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was still, no runs for which daughter product ratios were determined gave ratios of RaA to RaB to RaC greater than 3 to 1 to 0.4. Even with this departure from equilibrium, extrapolation to an infinite sample, assuming daughter product equilibrium, does not give a serious error. Thus it was not necessary to determine the relative daughter product concentration in order to calculate the theoretical dosage to the lungs from breathing for an extended period in the sampled atmosphere.

The experimental results are presented in chronological order. They show that the radioactivity of the rats' lungs was roughly proportional to the radioactivity removed by the filter paper from a liter of chamber air over a wide range of experimental conditions.

Table 14 covers the experiments performed with the first gamma counter discussed in Section 4.6. Table 15 covers the succeeding experiments.

Discussion of Table 14. Runs 1-6 were performed on three successive days. Outside air was blown into the chamber the previous night and in the early morning of the first day of the experiments. Runs 7-12 give the results of experiments in which the chamber was filled with filtered air. The walls of the chamber were lined with glycerine-coated glossy paper which, it was believed, might assist in removing dust from the air.

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The average gamma activity per unit radon concentration measured in the lungs of rats 1-6 which inhaled radon and its daughter products in atmospheric air that had not been cleaned was reduced 8.5 times in rats 7-9 by filtering the air before inserting it into the exposure chamber and 60 times in rats 10-12 by also fanning the air to further lower the air-borne daughter product activity. Cohn et al (8) reduced the daughter product activity measured in the lungs of rats by about 125 times by using an experimental arrangement in which rats were exposed to an air-radon mixture which was filtered of the daughter products just before being inhaled by the rats.

The results of runs 7-9 in Table 12 were obtained using filtered air which was not fanned. From the discussion in Section 6.3 we should expect that the reported RaC' levels, which were measured in the chamber proper, were several times higher than the RaC' concentration actually breathed by the rats in the more confined volume of their cages and cage holder. The increased retention to be expected under low dust conditions because of the increased mobility of the daughter products was not detected because of the overestimation of the RaC' levels breathed by the rats. The independently determined daughter product levels in a dummy cage under filtered air conditions were not used to correct the RaC' levels measured from the chamber in the filtered air experiments because it was uncertain to what extent the levels would be affected by the presence of the rat in the cage.

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It should also be noted that when the daughter products were not carried on dust particles, they may have been removed to a significant degree by the nasal passageways before reaching the lungs. Increased nasal deposition may thus have been partly responsible for failure to detect an increase in the per cent deposition in the lungs under low dust conditions. Measurement of deposition in the nasal passageways was not made in the present work.

Increased retention under low dust conditions was found in a human subject. These results are reported in Section 9.

Discussion of Table 15. Table 15 gives the results obtained with an improved counter. Run 13 was done with "normal atmospheric air, runs 14 and 15 in an atmosphere in which the dust had settled for sixteen days. We note that the introduction of the second rat increased the RaC' concentration considerably. This higher value was estimated from a ten minute sample. Run 16 was performed under stirred air conditions and produced results quite similar to the previous results under stirred conditions when filtered air had been used. In run 17, adding cigarette smoke to air whose dust load had been decreased considerably by settling, brought the RaC' concentration up tremendously with a corresponding rise of activity in the rats' lungs.

The glycerine-coated glossy paper, which had been placed on the walls before run 7, was removed from the walls of the chamber after run 17. In run 18, a cannulated dog was connected to the chamber at the same time the rat was in the chamber. The results of the dog experiments are reported in the next section. In this experiment, the RaC' concentration reached its highest

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level to that time under atmospheric air conditions, 81 per cent of equilibrium. Runs 19 and 20 give comparison readings between a rat in a settled atmosphere and one in a high dust atmosphere. The high dust atmosphere was obtained by blowing into the chamber finely ground ZnO_2 with an aerosolizer designed to disperse material in the solid phase. The powder increased the RaC' in the chamber air and increased the radioactivity of the rats' lungs proportionately.

7.3. Experiments with Restrained Rats.

In the experiments reported to this point, the rats were inserted into the chamber in cages. Their heads were often buried in their bodies while they rested in the cages and presumably this position could affect the amount of activity inhaled. To see if the rats' positions in fact made a difference runs 21 and 22 were done by inserting into the chamber two rats stretched out on rat boards. In this position it was impossible for the rats to contact their bodies with their nostrils. The rats were fairly calm during most of the run though at times they did struggle to free themselves.

In addition to the results given in Table 15 decay curves for the rats' lungs and the filter paper air sample are plotted in Fig. 11. The theoretical decay curves calculated on the assumption of daughter product equilibrium are compared with the experimental points. The results on the two rats were not significantly different from the results obtained with previous rats.

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Fig. 11. Curve (a). Measured decay of gamma activity in the lungs of two rats that had been exposed for several hours in an atmosphere containing radon and its daughter products. The curve presented in conjunction with the experimental data is a theoretical curve for an infinite sample of an atmosphere containing equilibrium concentrations of the daughter products.

Curve (b). Measured decay of alpha activity of a one liter grab air sample taken during the exposure of the rats. The curve presented in conjunction with the experimental data is a theoretical curve for a grab sample of an atmosphere containing equilibrium concentrations of the daughter products.

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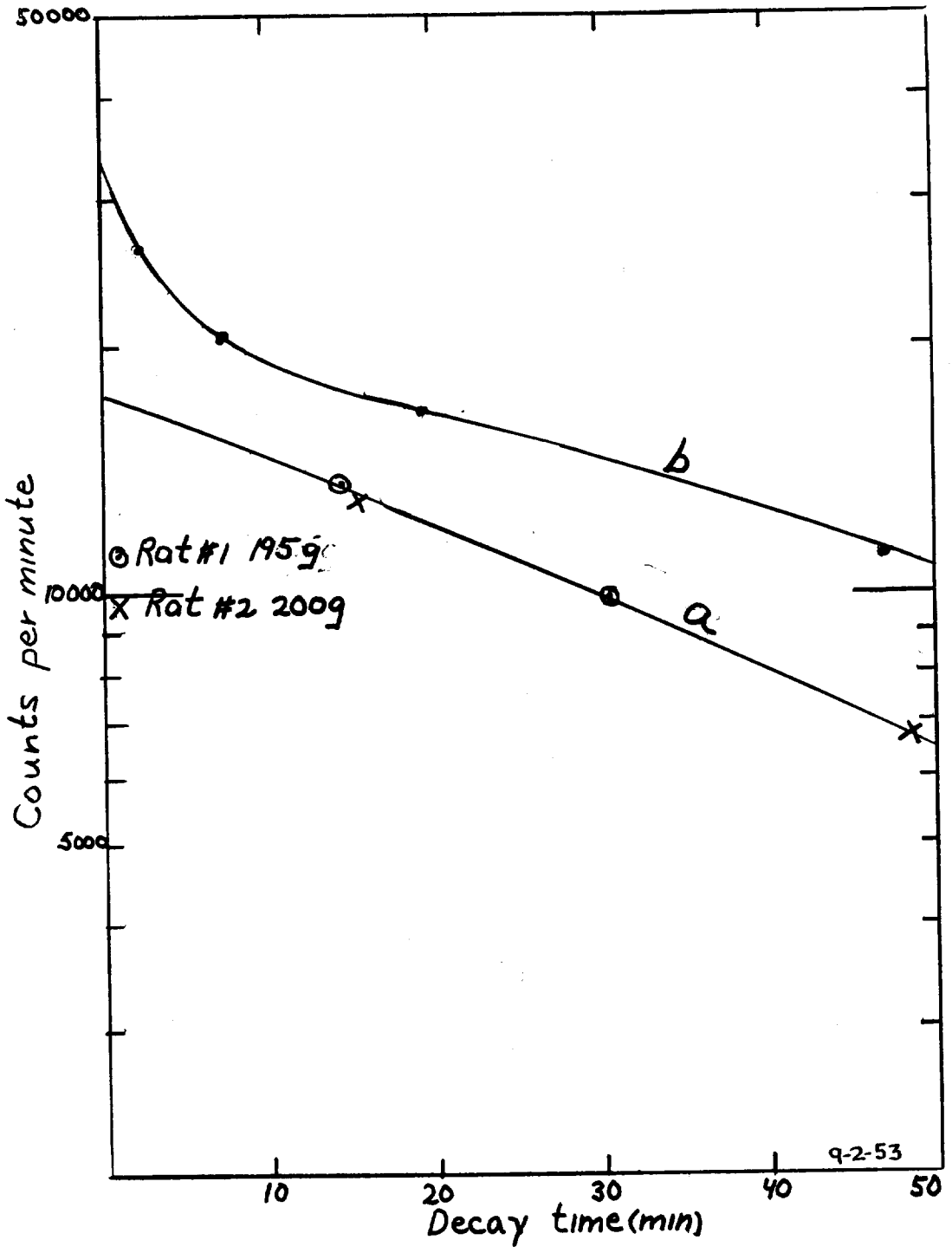

Fig. 11

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7.4. Dosages Received by Rats.

From the radiation hazard viewpoint, the only important dosage is from the alpha radiation. The total alpha dosage to the lungs can be obtained provided we know the efficiency of the gamma counter for the lung geometry, and the ratio of the alpha energy to the gamma radiation emitted in the lungs.

The efficiency of the counter used in runs 1-12 for different sample volumes and gamma energies was given in Section 4.4. The results, which were obtained for the radiations from Fe^{59} and I^{131}, are approximately applicable to the radiations from the radon daughter products.

Another way to obtain the efficiency of the gamma counter is from the number of gamma photons counted per alpha disintegration of a daughter product source. The true alpha disintegration rate of the daughter products removed from the air by millipore filters can be obtained readily from appropriate calibration experiments since self-absorption in the millipore source is small. Gamma counts are made on the same filter paper used for the alpha measurement and the alpha and gamma values are extrapolated to the same decay time. The measured γ/α ratio as compared to the theoretical ratio gives the efficiency.

The radon daughter product gamma spectrum was published by Evans (28) and is presented in Table 3. A total of 0.823 gamma quanta per disintegration of RaB and 1.455 gamma quanta per

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disintegration of RaC are given off. Using these values, theoretical gamma decay curves can be plotted. These curves are presented in Fig. 3 and 4.

Small errors in using the γ/α ratio method arise from differences in the energy spectra of the different samples counted and departures from daughter product equilibrium in the sampled atmosphere. The theoretical γ/α ratios for a grab sample remain fairly constant for a wide variation in relative daughter product concentrations. We see this in Table 16, which gives the γ/α ratios for instantaneous samples measured at specific decay times for the listed initial decay rates.

Table 16.

Number of Gamma Photons Emitted per Alpha Particle from Grab and Infinite Samples of Non-Equilibrium Daughter Product Atmospheres

Relative activities of daughter products in air			γ/α ratios Grab sample measured at			∞ sample
RaA	RaB	RaC	20m	40m	60m	No decay
10	10	10	2.02	1.91	1.84	1.84
20	10	6	2.19	2.00	1.89	1.81
30	10	4	2.30	2.06	1.92	1.76
10	0	0	2.38	2.23	2.00	1.14

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Thus the efficiency of Counter I may be determined by two independent methods:

Method 1. From Calibration with Fe^{59}.

The height of the rats' lungs in the test tubes was about two centimeters. A solution of Fe^{59}, 2.2 cm. deep, gave an efficiency of 9.7 per cent when the ore standard read 4270.

Assign an efficiency of 10 per cent for an ore standard reading of 4270.

Method 2. From γ/α Ratios.

We use the data from runs 1-6.

Run	α cpm	Time of α meas.(m)	γ cpm	Time of γ meas.(m)	α std.	γ std.
2	6200	88.5	1302	100	2080	4780
3	9160	53	1273	87	2100	5000
4	7300	62.5	850	101	2100	5000
5	2230	91	421	102	2133	5210
6	2780	80	493	97	2100	5180

We extrapolate the alpha and gamma measurements to t=0, extrapolate the gamma counts to a gamma standard of 4270, and convert the measured alpha counts per minute to total disintegrations per minute, using 4617 cpm = 100 per cent geometry for the alpha standard.

Run		Measured γ cpm / total α dpm
2		0.272
3		0.326
4		0.267
5		0.228
6		0.246
	Avg.	0.258

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The theoretical γ/α ratio at t=0 is 2.278. Therefore the efficiency is 0.258/2.278 = 11.3 per cent.

The filter paper occupied a volume of 0.5 cc. Measurements using the same volumes of Fe^{59} and I^{131} gave 12.3 per cent and 13.6 per cent respectively for the efficiencies.

Thus, efficiency determinations by two independent methods agreed fairly well.

Counter II was calibrated solely by the γ/α ratio method. The results are given in Table 17. The method of holding the filter paper in the test tube was changed for the measurements with Counter II. Previously the filter paper had been crumpled in the bottom of the tube so it filled about 0.5 cc. of the volume. An improved arrangement was obtained by filling the bottom centimeter of the tube with plaster of paris. The filter paper was then compressed against this base, thus occupying a more defined volume. Since the height was about one-half that occupied by the lungs, the efficiency of the counter for the lungs was approximately the same as for the filter paper.

Once the disintegrations per minute in the lungs has been determined, the ergs dissipated can be obtained if the average alpha energy per gamma photon is known. Table 18 gives the alpha energy per gamma photon emitted from the lungs for different non-equilibrium conditions in the inhaled air. We see from the table

1168666

Table 17.

Calibration of Counter II from γ/α Counts of Grab Sample

Date	γ/α counts	Sample decay time	Theor. γ/α ratio	Effic. %	Ore Std.	Effic. for Ore Std. II = 10000
8/1	0.314	108	1.74	18.05	9730	18.5
8/1	0.385	60	1.84	20.9	9730	21.5
9/2	0.354	60	1.84	19.3	10400	18.6
9/2	0.338	60	1.84	18.4	10400	17.7
					Average	19.1

Table 18

Ratios of Alpha Energy to Gamma Photons Given Off Simultaneously by Infinite Samples of Non-Equilibrium Daughter Product Atomspheres

Relative activities of daughter products in air			α Mev/photon
RaA	RaB	RaC	
10	10	10	4.12
20	10	6	4.12
30	10	4	4.18
10	0	0	6.0

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that an assumption of equilibrium will not lead to a serious error for the conditions that exist in practice.

Actually, the γ/α ratio of a grab sample should give a fairly close estimate of the γ/α ratio of an infinite sample, i.e. the lung, and thus give the lung dosage directly from the gamma measurement. This can be seen if we compare the theoretical γ/α ratios given in Table 16 for grab and infinite samples. The ratios are closest for a reading of the instantaneous sample taken after a decay time of sixty minutes and are equal if equilibrium conditions exist. Thus, the γ/α ratio of the instantaneous sample at sixty minutes can be used without further correction as the γ/α ratio of the infinite sample at zero decay time. The alpha energy imparted to the lungs in ergs can then be obtained from the average alpha energy per alpha disintegration, which is 7.58 Mev., and the relationship, 1 erg = 1.6×10^{-6} Mev.

If the weights of the rats' lungs are known, the ergs per gram can be obtained. Stokinger et al (34) found the average lung weight (wet) in the rat to be 0.43 per cent of the body weight. This formula has been used to calculate the results in Table 19. The relations 93 ergs/gm = 1 rep and 20 x rep = rem for alpha radiation have been used to obtain the rem in the lungs of the rats breathing radon and its decay products. The results are expressed in terms of mrem/hr per 10^{-9} curies/liter of RaC'.

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Table 19.

Dosages Received by the Lungs of Rats Breathing in an Atmosphere Containing Radon

Dosage from inhaled radon daughter products, calculated from activity measured in lungs

Run	Rat Wt.	Rn $c/\ell \times 10^9$	RaC' $c/\ell \times 10^9$	Ergs/hr	MREM/hr	MREM/hr. for 10^{-9} c/ℓ RaC'
1	189	24.1	12.7	8.18	2170	171
2	233	23.4	10.9	26.4	5650	518
3	197	18.8	8.07	8.15	2070	256
4	210	18.4	7.32	6.00	1430	195
5	190	15.5	4.08	5.15	1360	333
6	177	14.7	4.06	4.48	1270	313
7	175	25.2	1.22	1.41	402	329
8	198	24.9	1.69	1.50	378	224
9	170	24.9	1.27	1.32	388	306
10	184	20.2	0.272	0.19	51.7	190*
11	189	19.8	0.349	0.12	31.6	90.5*
12	180	16.3	0.116	0.14	38.8	334*
13	219	24.3	13.3	34.4	7880	593
14	215	22.8	2.68	4.97	1160	433
15	196	22.2	~7.5	10.1	2580	344*
16	189	16.5		0.17	45	
17	197	21.0	19.1	21.2	5400	282*
18	214	22.4	20.3	30.1	7030	346
19	247	22.6	7.61	10.8	2190	288
20	230	22.2	16.9	28.6	6200	367
					Average	334
					Std. dev.	118

*Omitted from averaging

Estimated dosage to lung of rat breathing 10^{-9} c/ℓ radon

from radon gas	2.58 MREM/hr.
from daughter products of radon molecules decaying in lung	6.45 MREM/hr.

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If we know the total volume of air in the rats' lungs, we can calculate the dosage produced in the lung by radon gas distributed through the lung in the same concentration as the inspired air and by the radon decay products in equilibrium with the radon. Guyton (33) gives the formula 0.0074 x weight of rat (gm) for the tidal volume of rats (cc.). Estimating the total lung volume as six times this, which is the ratio for humans, we have a rough idea of the dosage produced by radon and the daughter products in equilibrium with radon. This is compared with the measured daughter product dosage in Table 19.

7.5. Per Cent Retention of the Inhaled Daughter Products by Rats.

The per cent retention of the daughter products can be calculated if the minute volume is known. Guyton (33) reported that the average minute volume of slightly anesthetized rats as a function of rat weight was given by the formula:

Minute volume in cc/min = 0.65 x weight of rat in grams.

The theoretical gamma ratios for an infinite to a one liter grab sample are:

61.2 v for grab sample measured at 0 time

150 v for grab sample measured at 60 min.

where v is the minute volume in liters per minute.

Substituting Guyton's formula for the minute volume as a function of weight, v = 0.00065 wt., we get:

$$\frac{\text{Infinite sample counts (decay = 0)}}{\text{Inst. sample counts (decay = 60 min.)}} = 0.0975 \times \text{wt.}$$

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This ratio assumes radioactive equilibrium between radon and its daughter products.

Applying this formula to the runs in which gamma filter paper measurements were made we get the per cent retention values listed in Table 20.

Table 20.

Per Cent Retention of Daughter Products by the Rat from the Ratio of γ Counts of Lung to γ Counts of Filter Paper Air Sample

Run	Rat Wt.	F.P. counts at t= 60min.	Lung counts t=0	Meas. Lungs / F.P.	Theor. Lungs / F.P.	% Effic.
2	233	3100	7455	2.41	22.7	10.6
3	197	2280	2416	1.06	19.2	5.5
4	210	2040	1776	0.87	20.5	4.2
5	190	1028	1591	1.55	18.5	8.4
6	177	1100	1373	1.25	17.3	7.2
18	214	6240	14600	2.27	20.9	9.2
19	247	3160	5100	1.61	24	6.7
20	230	5980	13500	2.26	22.4	10.1
					Average	7.8
				Average	Corrected	9.2

In obtaining the efficiency of retention no attempt was made to take into account the relative efficiency of the counter for the lung and filter paper measurements. An additional error is introduced because of the difficulty in specifying accurately the daughter product concentration inspired by the rat. The measurements

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on the decay products given in Table 20 were made from the chamber. From the discussion of Section 7.1, we may expect that this was appreciably higher than the activity in the cage for very low dust conditions. Accordingly, it is possible to give a retention figure only for the daughter products carried primarily on atmospheric dust, that is, a figure obtained for rats breathing in air that was not cleaned. Filter paper samples obtained from the cage, using both slow and grab sampling, had activities about 15 per cent lower than the activities in the chamber. Using this correction figure, we calculate the average retention of the daughter products which were carried on atmospheric dust to be 9.2 per cent.

8. RESULTS OF DOG EXPERIMENTS.

8.1. Procedure.

To study the dosage of the radon daughter products to the lungs of dogs, the trachea of the anesthetized dog was cannulated and connected to the chamber through a respiratory check valve. The dog inspired air from the chamber through the valve whose outlet was connected through a gas meter back to the chamber. The gas meter was read periodically during the experiment to determine the expiredminute volume.

After it had been connected to the chamber for about two hours, the dog was disconnected and sacrificed. The lobes and

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trachea were dissected out individually, put into separate test tubes, and counted in the Texaco counter (see Section 4.4). In all the dog experiments outside air was blown into the chamber during the previous night and early morning of the day of the experiment. The chamber was then sealed and the radon inserted several hours before the dog was connected. The results are given in Table 21.

8.2. Determination of Respiratory Minute Volume.

Dogs 1, 2, and 4 were connected to the gas meter during the entire experiment. Dog 3 was connected directly to the chamber, thus eliminating the respiratory check valve. Its minute volume was measured with the gas meter at the beginning of the experiment. At the end of the experiment its minute volume was estimated very roughly by considering it proportional to the respiratory rate. No measurements were made on the respiration of dog 5.

The respiratory data on the dogs are given in Table 22. The average minute volume was obtained by multiplying each determination by a decay factor obtained from the gamma decay curve of the daughters starting in equilibrium. The decay time was chosen from the middle of the measurement period for a given determination to the time the dog was disconnected. The readings were also weightedproportionally to the time between the successive readings of the gas meter.

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Table 21.

Results of Dog Experiments

	Dog No. 1	2	3	4	5
Weight(kg.)	13.3	8.35	9.54	9.6	8.48
Minute volume (ℓ/m)	3.1	2.8	1.7		0.35
α/γ ratio at t = 60m.	15.6	19.1	19.1	19.7	19.7
RaC' concentration (c/ℓ x 10^9)	19.2	21.2	21.2	18	18
γ cpm at t = 0					
Lobe A	11320	11600	6950	5610	4360
Lobe B	5280	4760	2400	3345	2835
Lobe C	13370	10300	8520	7970	5170
Lobe D	5050	3410	3720	4020	2500
Lobe E	4940	6310	2500	2840	2195
Lobe F	12750	11080	8525	10690	4930
Lobe M	5840	3630	3040	5430	2245
Activity of lung	58550	51090	35655	39905	24235
Trachea	1070	1570		1790	204
Lobe weights(gm.)					
Lobe A	16.0	11.5	11.5	14.5	12.5
Lobe B	11.0	6.0	4.5	8.25	8.0
Lobe C	28.5	18.0	16.5	19.75	20.0
Lobe D	9.0	4.0	6.5	7.25	8.8
Lobe E	6.5	7.5	5.0	4.5	5.5
Lobe F	30.5	17.5	16.0	23.75	20.0
Lobe M	9.5	5.5	4.5	7.75	6.5
Weight of lung	111	70	64.5	85.75	81.3
Trachea - irradiated weight	0.23	0.23	0.23	0.23	0.23

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Table 22.

Minute Volume Measurements in Dog Experiments

I Time min.	II Resp. m^{-1}	III Meter ft^3	IV ℓ/min.	V Decay Time	VI Decay Factor	VII Rel. time between readings	VIII VI times VII	IX Product of IV, VI and VII
Dog No. 1								
0		75.4						
18		76.35	1.49	173	0.032	1.2	0.038	0.057
41	10	77.24	1.09	150	0.054	1.53	0.083	0.090
56		78.31	2.05	135	0.076	1	0.076	0.16
71	12	79.81	2.82	120	0.11	1	0.11	0.31
86	12	81.13	2.48	105	0.15	1	0.154	0.38
101	14	82.47	2.52	90	0.22	1	0.219	0.55
116	17	84.45	3.72	75	0.30	1	0.30	1.12
131	20	86.13	3.16	60	0.41	1	0.41	1.30
146	20	88.07	3.65	45	0.55	1	0.55	2.01
159	22	89.47	3.06	30	0.71	0.87	0.62	1.9
183	Dog disconnected						2.56	7.88
							Average = 3.08 ℓ/m	
Dog No. 2								
0	8	3.33						
15	8	4.25	1.74	80	0.26	1	0.256	0.445
30	10	5.38	2.13	65	0.36	1	0.356	0.758
45	13	6.40	1.92	50	0.48	1	0.48	0.92
71	34	9.66	3.64	31	0.69	1.73	1.19	4.33
88	Dog disconnected						2.28	6.45
							Average = 2.82 ℓ/m	
Dog No. 5								
0	~1	0						
18		0.23	0.36					
33		0.4	0.32					
54		0.64	0.32					
79		0.96	0.35					
98		1.20	0.36					
104		1.28	0.37					
116	Dog disconnected Chose 0.35 ℓ/m = min. vol.							

Dog No. 3

At t = 0, min. vol. = 1.1 ℓ, respiration = 10/m

t = 180m , dog disconnected, Respiration = 15/m

Estimated average min. vol. = 1.7

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8.3. Correction of Counts for Geometry.

The counting rates of the lobes were corrected for differences in counting efficiences caused by the different heights of the lobes in the test tubes. The correction was minimized by locating the lobes at that portion of the counter where the variation with sample volume was least. Corks were inserted into the test tubes to bring the bottoms of the lobes approximately 2.5 cm. from the bottom ends of the test tubes. To determine the correction, a disk source, prepared from a filter paper sample of the daughter products, was counted at various heights in a sample test tube filled with water. The maximum correction was about 5 per cent.

8.4. Sampling of Air for Daughter Products.

The daughter products were sampled from the chamber next to the place where the respiratory check valve leading to the dog was connected. An independent experiment was performed to determine the loss of radioactive material in the dead space (about 30 cc.) of the valve-tracheal cannula system. Several air samples with the water aspirator were taken alternately from the end of the tracheal cannula and directly from the chamber. The sample volumes were about 150 cc. First, about 200 cc. of air from the cahmber was flushed through the cannula. After a

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delay of ten seconds to simulate the delay between inspirations, the 150 cc. filter paper sample was drawn. The results of this experiment are given in Table 23. Since the time between collection of the sample and its measurement was the same for each sample, no correction for decay was necessary.

Table 23

Extent of Daughter Product Loss in Tubes Connecting Dog to Chamber

Sample No.	Sample Volume ml.	Counts	Counts for 120 cc.
Sample collected through cannula and valve			
1	160	2739	2055
2	150	2456	1960
3	117	2053	2105
4	119	2132	2150
5	126	2088	1990
		Average	2052
Sample collected directly from chamber			
6	118	2208	2246
7	119	1952	1970
8	121	2261	2240
9	122	2268	2230
		Average	2172

The average activity for the air samples drawn directly from the chamber was 6 per cent greater than the average for the sample drawn through the cannula. The retention figures reported in Table 24 were increased by 6 per cent over the

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measured values to correct for this difference. Using this correction an average retention of 11 per cent was found for the dogs with the higher respiratory rates and a retention of 50 per cent was found for the dog that was deeply anesthetized.

Table 24

Per Cent of Inhaled Daughter Products Retained by the Dog.

Dog No.	γ cpm lungs	γ cpm F.P.[a]	I Lung cpm / F.P. cpm	Min. vol. ℓ/m	II 150MV	% Ret.[b] I & II
1	58550	1200	48.9	3.08	465	11.1
2	51090	1230	41.5	2.82	426	10.3
3	35655	1230	29	1.7	255	12.1
5	25760	1030	25	0.35	52.5	50.3

[a] γ activity of liter grab sample, counted after one hour decay.
[b] Corrected for daughter product loss in tracheal cannula and respiratory check valve.

8.5. Dosages Received by Dogs.

Table 25 lists the calculated dosages to the lobes and the trachae of the five dogs. The dosages are extrapolated to a common equilibrium daughter product concentration of 10^{-9} c/ℓ. The lobes had been dried overnight in an oven at 90° C. before weighing, so the weights were multiplied by five (35) to give the approximate wet weights in the living animal. Although the minute volume of dog 5 was only 14 per cent of the average minute volume of dogs 1-4, the dosage to the lungs was 62 per cent of the

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Table 25.

Dosages Received by the Lungs of Dogs Breathing in an Atmosphere Containing Radon

	Dog No. 1	2	3	4	5
Minute volume (ℓ/m)	3.1	2.8	1.7		0.35
Respiration (m^{-1})	20	30	15		~1
Ergs/hr					
RaC' ($c/\ell \times 10^9$)	19.2	21.2	21.2	18	18
Lobe A	128.4	161.4	96.6	80.5	62.5
Lobe B	59.8	66	33.6	48.0	40.6
Lobe C	151.4	142.8	118.2	114.3	74.1
Lobe D	57.3	47.4	51.6	57.6	35.8
Lobe E	56.0	87.6	34.8	40.7	31.5
Lobe F	144.6	153.6	118.2	153.1	70.8
Lobe M	66.2	50.4	42.6	77.9	32.2
Ergs/hr to lung	663.7	709.2	495.6	572.1	347.5
Trachea	12.2	21.8		25.6	2.92
MREM/hr for 10^{-9} c/ℓ RaC'					
Lobe A	89.9	143	85.3	66.3	59.7
Lobe B	60.8	112	75.9	69.5	60.6
Lobe C	59.5	80.5	72.8	69.0	44.3
Lobe D	71.4	120.5	80.6	95.0	48.6
Lobe E	96.5	118.8	70.6	108.0	68.5
Lobe F	53.0	89.1	75.2	77.0	42.2
Lobe M	78.0	93.2	96.2	120.0	59.3
Average over lung	67	102.7	78.0	79.6	51.1
Trachea	580	950		1300	167

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average dosage for the other four dogs. This slight dependence of dosage on minute volume points to a diffusional process as the main mechanism for deposition of the daughter products.

An interesting result is the large dosage of 940 mrem/hr /10^{-9}c /μ RaC' to the trachea. This is over ten times the average dosage imparted to the lobes of the lung. The dosage to the trachea was obtained by assuming the alpha energy was dissipated in a cylindrical shell, 66 mu thick. This is the range in water of alpha particles with the average energy of the alpha particles emitted by the daughter products that were removed from the inhaled air. The inner diameter of the shell was taken as 1 cm. and the length as 11 cm. These were approximately the dimensions of the tracheae whose activities were measured.

9. DAUGHTER PRODUCT RETENTION IN MAN.

9.1. Procedure.

The fraction of the inhaled radon daughter products retained in the lungs of a human subject was measured by using an apparatus which appreciably decreased the effort required to breathe through a millipore filter. The experimental arrangement is shown schematically in Fig. 12.

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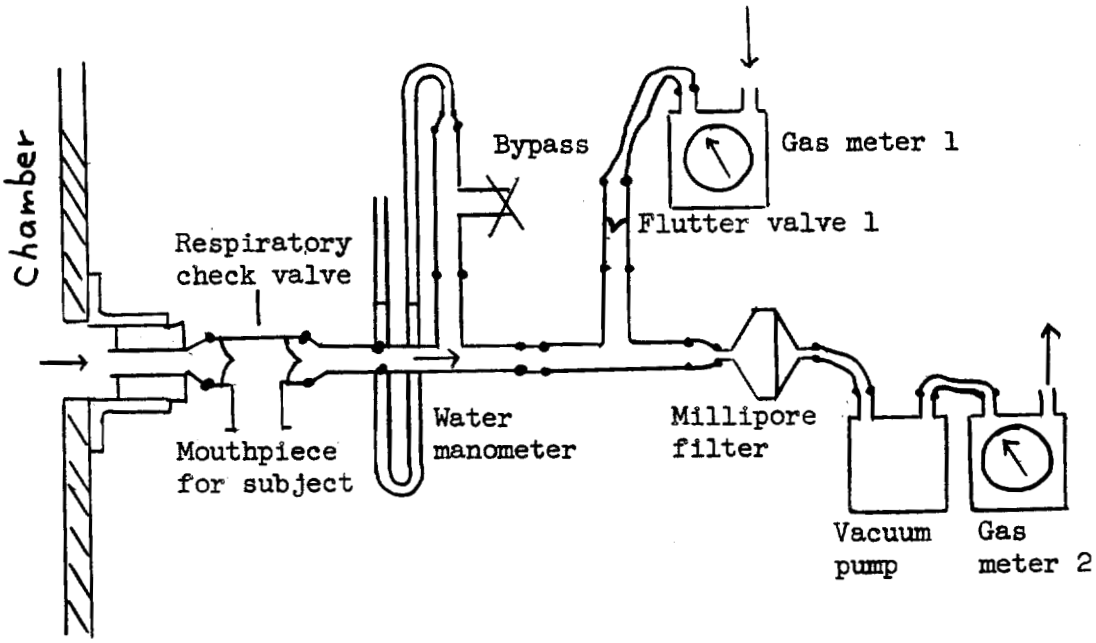

Fig. 12. Experimental arrangement for human inhalation experiment.

The vacuum pump in Fig. 12 operated during the entire run. When the subject did not expire through the check valve, room air was drawn through gas meter and inspiratory valve 1 and through the millipore filter. When the subject exhaled, flutter valve 1 was closed and the subject's air was drawn through the millipore filter by the vacuum pump. The pressure at the outlet end of the respiratory check valve was about -1 cm. H_2O because of the flow of air

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through meter 1. The chamber was maintained at about -3 cm. to insure that no air would be drawn by the vacuum pump directly from the chamber. In a control experiment, the outlet of the check valve to the lung was stoppered and the pump was turned on for several minutes. Negligible activity was measured on the filter paper.

The procedure for a run was as follows:

1. The outlet of the respiratory check valve to the subject was stoppered. The pressure in the chamber was made equal to atmospheric pressure. The connection to valve 1 was pinched off. About fifteen liters of air were drawn from the chamber through the millipore filter and gas meter 2 by the vacuum pump. This brought the pressure in the chamber down to about -10 cm.

2. The chamber pressure was brought back to -3 cm. A new filter paper was quickly inserted into the millipore holder. The subject then began breathing from the chamber. His first ten exhalations were bypassed around the filter paper to equilibrate him with the chamber. About fifteen exhalations were then passed through the millipore filter. During the experiment the air in the chamber was maintained at about -3 cm.

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3. After the subject had completed breathing, a new millipore was inserted and the air sampled again as in 1.

The activity on each millipore filter was left to decay the same length of time before counting, to eliminate the need to make corrections for decay.

The volume exhaled by the subject was obtained from the difference in the readings between meter 1 and meter 2. The zero reading was obtained from the readings of the two meters when air was passed through them in series.

The experimental results were converted to counts per minute per liter of air through the millipore filter. The ratio between the activity of the expired air sample and the sample drawn directly from the chamber gave the fraction of the daughter products that was not retained.

9.2. Results.

In the first series of experiments the retention was measured using air which had been freshly inserted into the chamber from the outside. In the second series of experiments filtered air was used. The results are given in Tables 26 and 27.

The average retention of the daughter products in air containing normal atmospheric dust was 25 per cent. The average retention of the daughter products in filtered air was

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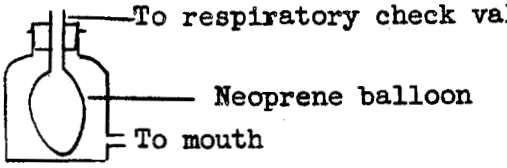
Table 26.

Per Cent Retention of Daughter Products Carried on Atmospheric Dust from Human Inhalation Experiment

	Counts/ℓ			% Retention	
	I Air Sample A	II Exhaled air	III Air Sample B	From II & I	From II & III
Run 1					
1st reading	673	465	609	31	23.6
2nd reading	658	455	596	30.9	23.7
Run 2					
1st reading	1065	811	1040	23.8	22
2nd reading	1062	802	1012	24.5	21.9
Run 3					
1st reading	49.7	37	49.2	25.5	24.7
2nd reading	49.3	37.2	47.6	24.7	22
Artifical lung[a]					
1st reading	49.2	48.9	47.6		
2nd reading	47.6	48.5	46.7	...	...

	Run 1	Run 2	Run 3	Art. lung
Radon($c/\ell \times 10^9$)	2.92	2.92	1.06	
RaC' ($c/\ell \times 10^9$)	2.14	2.14	0.68	
Min. vol. (ℓ/m)	11		9.4	
Tidal vol. (ℓ)	1.3	1.3	1.1	
Sample A (liters)	14.2	14.3	14.4	14.4
Exhaled air (liters)	38.4	18.9	16.4	18.2
Sample B (liters)	14.2	18.5	14.4	14.4

[a]The artificial lung was a neoprene balloon which was enclosed in a jar.



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75 per cent. The minute volume was ten liters and the tidal volume was over one liter.

Table 27

Per Cent Retention of Daughter Products in Cleaned Air from Human Inhalation Experiment

	Counts per liter			Exhaled Activity	%
	Air Sample A	Exhaled air	Air Sample B	Avg. A and B	Retent.
Run 1	193	47.5	151	0.28	72
Run 2	359	72.7	289	0.22	78

Radon (c/ℓ x10⁹)	9.3
RaC' (c/ℓ x10⁹)	1.0
Tidal vol. (ℓ)	~1.5

	Run 1	Run 2
Sample A (liters)	8.88	11.38
Exhaled Air (liters)	15.74	16.63
Sample B (liters)	8.52	11.38

These retention figures may be compared to the 50 per cent retention figure reported by other investigators who made the measurements using radon mixed with room air (36, 37). Though it is difficult to compare the results obtained for different subjects under unknown breathing conditions, it may be expected that the room air was in a state somewhat intermediate between the two extremes which were employed in the chamber investigations. The larger size of the room, along with the lower dust load in the air, favored a higher concentration of the more mobile daughter products than were present in those chamber experiments conducted shortly after the outside air was inserted. This would give a higher retention figure for

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the room experiments except when the chamber air was cleaned of dust.

Using these values we can calculate the dosage from continuous inhalation of the radon daughter products averaged over the entire lung. For a RaA-RaB-RaC-C' concentration of 10^{-9} curies/liter, 308 ergs/hr alpha energy would be delivered to the lung at 25 per cent retention. This amounts to 3.3 mrep/hr or 66 mrem/hr if we assume the lung weighs one kilogram. Using the ratio of 10 to 1 for the dosage in the trachea to the rest of the lung, obtained from the dog experiment, we estimate the dosage to the trachea as 660 mrem/hr. These results are for daughter products carried on atmospheric dust. Similar calculations can be made for the dosage from inhaling the daughter products that are air-borne in cleaned air. We should expect the trachea to receive a relatively higher dosage because of the greater mobility of the daughter products in cleaned air. An additional uncertainty in the calculations for cleaned air is introduced by lack of knowledge on deposition in the nasal passageways under these conditions. When the daughter products are suspended in filtered air, this deposition may be significant.

These calculated dosages are based on the assumption that the deposited daughter products remain at the site of deposition until they decay. Wilson and LaMer (38), working with a soluble Na(24)Cl aerosol, found that the lung radioactivity of human subjects decreased by one-third about fifteen minutes

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after cessation of inhalation. Stokinger et al (34), working with insoluble particulates, did not find any significant removal over short periods of time from the alveolar regions of rats' lungs.

The retention of the daughter products after a period of several hours probably varies in different regions of the lung. The ciliated regions, which extend down to the bronchioles, may remove the daughters more readily than the non-ciliated regions. However, for lack of any quantitative information, it was assumed in all the calculations that the daughter products remained where they had been deposited until they decayed.

10. CALCULATIONS OF THE RADIATION DOSAGE TO THE LUNGS OF MAN BREATHING IN AIR CONTAINING RADON.

The experimental figure for human retention of the radon daughter products obtained in Section 9 was used to calculate an average dosage to the lung. We should have a more realistic idea of the radiation hazard if we could assess the dosages imparted to the different regions of the lung. These dosages depend on the pattern of deposition of the particulate matter which carries the daughter products. We shall attempt to calculate the dosages to the constituent regions of the lungs for several sizes of particles on which the daughter products are likely to be deposited.

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10.1. Particle Sizes Selected for Dosage Calculations.

In normal atmospheric air, the radon daughter products exist largely on nuclei whose diameters vary between 0.004 mu and 0.12 mu (24, 39). The size of the nuclei seems to depend on the humidity. These nuclei are produced by spray from the ocean and by combustion in industrial areas. Their concentrations have been measured as high as 250000/cc. in city air and as low as 7/cc. in clean country air (40).

In a mine, when the air is still, we should expect only a small fraction of the daughter products to be on nuclei. The rest either exist as suspended matter of high mobility or become attached to the walls. However, when the air is dusty, the radon daughter products become attached to the dust and produce a spectrum of radioactive dust. The radioactivity of each size is proportional to the combined surface area of all the particles of that size. The size frequencies of dusts produced by drilling and blasting sandstone are given in Table 28.

The available experimental data indicates that few particles less than 0.2 mu in diameter are formed in mineral dusts produced by mechanical disintegration processes (41). Gibbs (42) concluded that the lowest theoretical size to which solids or liquids can be reduced in size by disintegration methods is of the order of 0.1 mu. This limit is imposed by the operation of

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Table 28.

Size Frequency of Particles in Various Mineral Dusts

from H. L. Green, Trans. Faraday Soc. 32, 1091, (1936)

Size Group diam. (mu)	Per cent Frequency	
	Shale during drilling	Shale after blasting
Up to 0.2	7.6	10.0
0.2 - 0.4	13.6	10.5
0.4 - 0.8	15.6	17.0
0.8 - 1.2	10.6	12.5
1.2 - 1.6	9.3	19.5
1.6 - 2	8.0	6.5
2 - 2.5	7.6	4.0
2.5 - 3	7.0	10.0
3 - 4	6.6	6.5
4 - 5	3.6	4.6
5 - 10	3.0	4.0
Above 10	7.5	5.0

Samples were obtained with the thermal precipitator. They were examined with a microscope fitted with a 2mm oil immersion apochromatic microscope objective and a x17 eyepiece giving 1500 diameters magnification.

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cohesional or surface tension forces upon each separate fragment of material as it is being formed during the process of disintegration. Green (41) reported that the appearance of smaller size particles in mine dust was shown to be produced from smoke particles derived from explosives used in disrupting the rocks.

It is instructive to calculate the dosage imparted to the lungs by those radioactive particles which may exist in significant amounts in mine atmospheres (i.e. 0.2 mu to 6 mu diameter) and by particles which normally exist in the air. We shall select a particle with a diffusion coefficient of 16×10^{-6} cm.^{2}/sec. (approx. 0.06 mu diameter) as typical of the permanent atmospheric nuclei (43). This size is characteristic of the large Langevin ions in the air. Most of the air-borne radioactivity appears to have been measured on the so-called "intermediate" nuclei originally discovered by Pollack (44) which are more mobile. The selection of the larger particle size seems justified, however, in view of the discovery by Pollack that the intermediate ions disappear presumably to give large ions, in regions of high humidity. The transformation to the larger size then should occur in the saturated atmosphere of the human respiratory tract. In addition, we shall calculate dosages from inhaling the daughter products as positive ions [diffusion coefficient = 0.03 cm.^{2}/sec (45).]

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10.2. Review of Published Experimental Data on Deposition of Particulate Matter in the Human Respiratory Tract.

Several extensive investigations of the deposition of particulate matter in the human respiratory tract have been reported (46). Some of the results are given in Fig. 13. Van Wijk and Patterson (47) carried out experiments underground in a South African gold mine in which the rock consisted mainly of quartzite. Using an optical microscope, they made a size analysis of dust which was collected from inhaled and exhaled air with the thermal precipitator. They used three subjects with an average minute volume of 17 liters and an average breathing rate of 19.

Wilson and LaMer (38) used a radioactive monodisperse aerosol of particles consisting of equal volumes of water and glycerol and a small amount of NaCl containing a trace of Na^{24}, to obtain retention data as a function of the breathing rate for seven human subjects. The inhaled and exhaled air were analyzed for radioactivity and the retention was calculated from the difference. In addition, Wilson and LaMer counted the gamma activity in the lungs by placing a GM tube directly against the chest. They used an experimental arrangement which they believed gave them mainly alveolar counts. They found a maximum alveolar retention for particle diameters between 0.7 mu and 1.7 mu.

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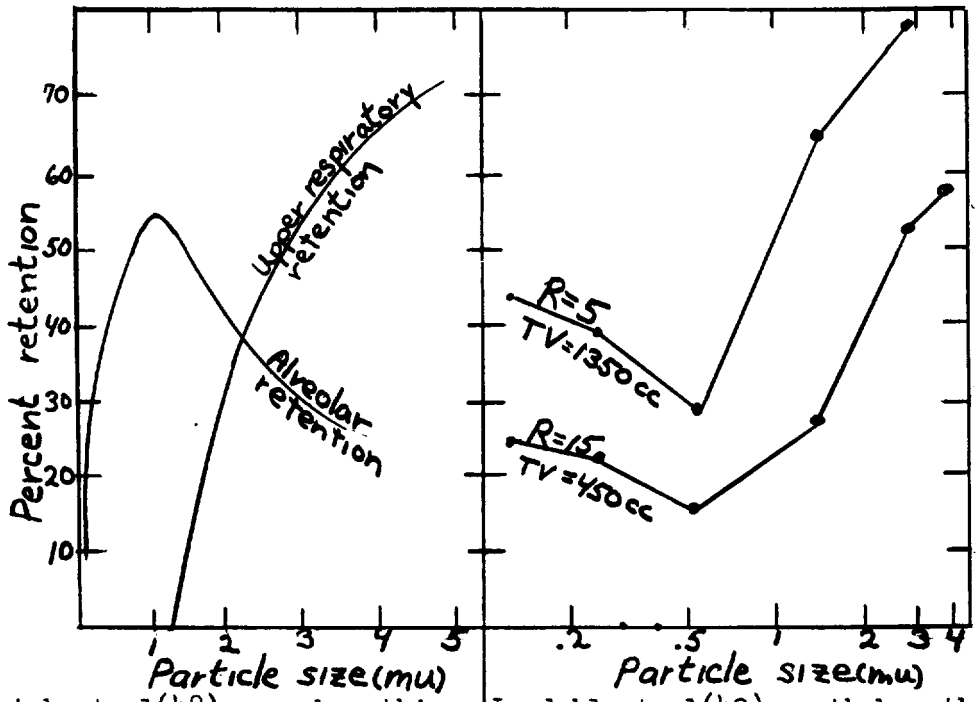
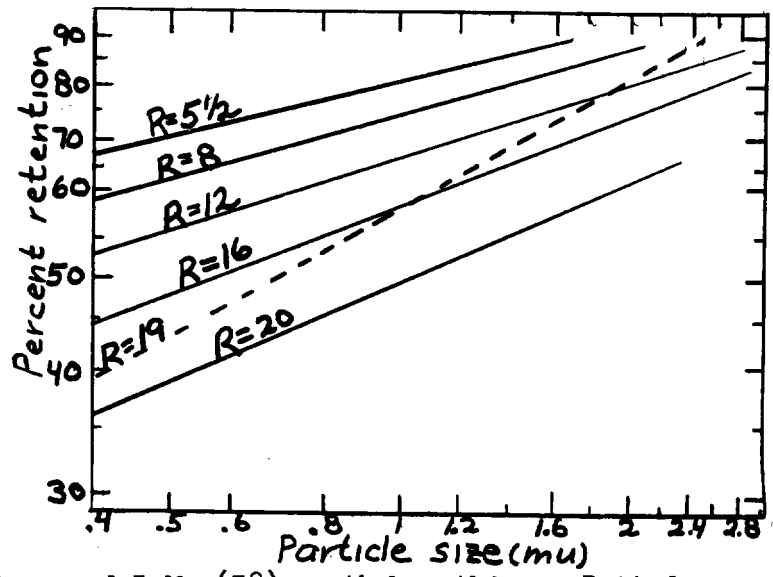


Hatch et al(48)-nose breathing | Landahl et al(49)-mouth breathing



Wilson and LaMer(38)-mouth breathing. Dotted curve gives data of Van Wijk and Patterson(47) for nose breathing.

Fig. 13. Published curves of percent retention of dust particles by human subjects as a function of particle diameter in microns. R=respirations/min. TV=tidal volume.

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They presented arguments which indicated that the maximum alveolar retention was of the order of 45 per cent of the inhaled aerosol.

Hatch et al (48) measured the retention of china clay dust, density 2.6, as a function of particle size. The size range was from 0.2 mu to 5 mu. Their results were in fairly close agreement with that of Wilson and LaMer, in spite of the difference in density of the material used. They collected the exhaled air in fractions and classified the total retention into alveolar and upper respiratory fractions. The maximum alveolar retention was for particles with diameters a little larger than one micron.

Landahl et al (49) carried out experiments with aerosols of homogeneous droplets of triphenyl phosphate for several particle sizes. Data was obtained for twenty-four subjects, employing several different breathing patterns. A minimum retention was found at 0.5 mu diameter. An average retention of 41 per cent was found at 0.25 mu diameter for a 1350 cc. tidal volume.

10.3. Deposition of Particulate Matter in the Human Respiratory Tract - Theoretical.

Calculations of the deposition in the lungs of particles with diameters between 0.2 mu and 6 mu were made by Landahl (50). He used a model of the respiratory tract patterned after one

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originally proposed by Findeisen (51). The dimensions of the model are given in Table A.1 of the appendix. Air inhaled into the tree passes through several different regions in succession. Each region consists of a number of identical tubes which branch in turn into another set of identical tubes which comprise the next region. The particles are removed from the air stream by settling to the bottom of the tube under the influence of gravity, by diffusing to the sides of the tube as a result of their Brownian movement, and by impacting against the walls of bends in the tube because of their inertia. In accounting for the total per cent retained, impaction is of importance for the particles with diameters larger than 3 mu, diffusion and sedimentation for particles between 0.2 mu and 1 mu, and diffusion is the most important removal process for the smallest particles.

The smallest particle size considered by Landahl was 0.2 mu. His results are given in Table 29.

The theoretical analysis of the deposition of particulate matter was extended in this investigation to include calculations of the deposition of particles0.06 mu in diameter and of positive ions, since most of the air-borne daughter product activity is carried on particles between these extreme sizes. The only important process for the removal of these particles is

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Table 29.

Per Cent Deposition of Inhaled Particles in the Constituent Regions of the Human Respiratory Tract

Tidal volume = 450 cc.
Minute volume = 6.75 ℓ/m

partly from H. D. Landahl, Bull. Math. Biophys, 12, 43 (1950)

Region	Diameter					
					Sub-microscopic	
	6u	2u	0.6u	0.2u	$D=16\times10^{-6}$ [a]	$D=0.03$ [b]
1. Mouth	0	0	0	0	0.027	5.2
2. Pharynx	0	0	0	0	0.013	2.6
3. Trachea	1	0	0	0	0.033	6.0
4. Prim. Bronchi	2	0	0	0	0.033	4.8
5. Sec. B.	4	0	0	0	0.075	8.5
6. Ter. B.	9	0	0	0	0.23	17.4
7. Quart. B.	7	1	0	0	0.43	19.8
8. Terminal B.	19	4	2	2	6.4	22.6
9. Resp. B.	11	4	2	2	6	
10. Alv. Ducts I	25	16	5	6	16.4	
11. Alv. Ducts II	5	28	10	9	15.3	
12. Alv. Sacs	5	0	0	0		
	83	51	19	19	45	87

[a] Diffusion coefficient (cm^2/sec) of atmospheric condensation nuclei.
[b] Diffusion coefficient of positive ions.

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diffusion. The basis of the calculations is an equation for the deposition by diffusion of the particles flowing in a cylindrical tube derived by Gormley (52). A sample calculation is given in Appendix 2. The value of the diffusion coefficient of the atmospheric nuclei is that measured by Nolan (43) for the condensation nuclei in atmospheric air stored for 1.7 hours. The effect of mixing is neglected in the calculations. Mixing increases the number of particles removed from the air stream.

Variables in addition to particle size which affect deposition in the human respiratory tract are particle density, depth of breathing, rate of breathing, and factors which may enter to change the particle as it passes through the tract. Hygroscopic particles are likely to increase appreciably in size as they enter the saturated atmosphere of the respiratory tract.

The calculations are made for a 450 cc. tidal volume and a 4 second respiratory cycle. This gives 15 breaths per minute. The cycle is divided into the following fractions: 3/8-inspiration, 1/8 pause, 3/8 expiration, 1/8 pause. Landahl's results apply to particles of unit density. They may be extended to particles of different density, ρ, if they are larger than one micron in diameter by assigning them an equivalent diameter equal to $\sqrt{\rho}\, d$.

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10.4. Comparison of Theoretical and Experimental Results.

The available experimental data agrees fairly well with the theoretical calculations down to 0.2 mu (49). The experimental data is meager below 0.2 mu because of the difficulty of determining smaller particle sizes.

It is difficult to compare experimental values of the deposition of the radon daughter products obtained in this investigation with theoretical calculations because of the uncertainty of the value of the diffusion coefficient for the air-borne radioactivity. The diffusion coefficient of 16×10^{-6} cm ^{2}/sec used to obtain the results in Table 29 is the largest value for atmospheric condensation nuclei reported by Nolan. Nolan also reported $D=9.6 \times 10^{-6}$ for nuclei which had been stored for 2 hours and $D = 2.8 \times 10^{-6}$ for air stored 39 hours. Even if we use the smallest value of the diffusion coefficient given by Nolan, we still calculate a deposition of about 60 per cent for the breathing pattern employed by the human subject in Section 9, as compared to the measured value of 25 per cent. The explanation of this difference must await further investigation of the suitability of the idealized model of the respiratory tract and the real nature of the air-borne carriers of the daughter products.

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Although the model may not give the total retention correctly, it should be more successful in giving relative dosages to the different sized bronchial tubes in the lung.

10.5. Calculation of the Dosages to the Constituent Regions of the Respiratory Tract.

Dosages to the constituent regions of the respiratory tract were calculated from the average alpha-particle energy expended per gram in cylindrical shells whose inner diameters equal the inner diameters of the passageways in the tract and whose thicknesses equal the range in water of alpha-particles with the average energy of the alpha particles absorbed in the tissue. Table 30 gives the rem in the different regions of the respiratory tract for the following two cases:

a) inhaling a radon concentration of 10^{-9} curies per liter and assuming radon is in equilibrium with its daughter products in the respiratory tract.

b) inhaling an equilibrium RaA-RaB-RaC-C' concentration of 10^{-9} curies per liter. The dosages were calculated for the carrier-particle sizes and deposition data given in Table 29.

The dosages apply only to the breathing pattern given in Table 29. For breathing that is deeper than that assumed in the calculations, the per cent deposition of inhaled particles is much higher, but the increased deposition is all in the

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Table 30

Calculated Dosages to the Constituent Regions of the Human Respiratory Tract from Inhaling 10-9 Curies Per Liter Radon in Equilibrium with Its Daughters

	MREM PER HOUR							
	100% dep. in region[a]	6mu	2mu	0.6mu	0.2mu	D = 16×10^{-6}	D = 0.03	Radon[b]
Trachea	4.46×10^5	4500	0	0	0	147	26800	70
Prim. Br.	7.18×10^5	14000	0	0	0	237	34500	44
Sec. Br.	5.83×10^5	23000	0	0	0	437	49500	18
Ter. Br.	2.76×10^5	25000	0	0	0	635	48000	8.8
Quart. Br.	1.43×10^5	100000	1430	0	0	615	28300	6.4
Term. Br.	7200	1370	287	144	149	460	1630	2.4
Resp. Br.	8100	890	322	161	161	485		1.3
Alv. D. I	1550	380	240	78	93	246		0.97
Alv. D. II	222							0.78
Alv. Sacs	63.6							
Dosage averaged over 1kg. lung	177	147	90	34	34	80	154	2.6

[a]Assumes all inhaled daughter products deposited in specified region, = $1.78 \times 10^5 \div$ volume irradiated by alpha particles.

[b]Dosage from inhaled radon and daughter products of radon molecules decaying in lung, = 0.88x volume of radon ÷ volume irradiated by alpha particles.

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alveolar region. Because of the large volume that is available to absorb the alpha energy in the alveolar region, the resulting dosage to alveolar tissue is still relatively small. Thus, the increased deposition at deeper breathing is probably not very significant from a health physics viewpoint.

If the breathing rate is doubled, but the depth of breathing remains the same, the per cent deposition per respiratory cycle will decrease because of the shorter time spent by the Brownian particles in any particular region. Calculations made for a flow rate of 600 cc /sec (Min. vol. = 13.5 ℓ/m) give a deposition of 32 per cent compared to a deposition of 45 per cent for the minute volume of 6.75 ℓ/m.

The most striking result of the calculations is the variation in dosages imparted to the different regions of the respiratory tract. Table 31 gives the ratio of the maximum regional dosage for a given particle size to the dosage averaged over the whole lung. For the particles on which almost all the radioactivity is deposited, that is, between 2 mu and 0.03 mu, the ratio varies between 5 and 16 times the average dosage. The maximum dosage is 320 times the average for positive ions and 670 times the average for 6 mu particles.

The dosages at the branch points in the respiratory tract were not considered. Dosages at these regions could conceivably be many times higher than the average dosages to the other regions of the tract.

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The model used in the calculations presented in this section was based on mouth breathing. Deposition in the nasal passages is negligible except for the largest particles and for the positive ions, which should generally account for only a small fraction of the daughter product activity. Thus the calculated lung dosages are equally valid for nasal breathing and mouth breathing when the daughter products are carried on nuclei normally present in the air. Instances are conceivable, however, when the daughter products may exist to a large extent as highly mobile ions or molecular aggregates in the air. These may be deposited to a significant degree in the nasal passageways. However, the writer does not have enough information at the present time to assess the role of the nasal passageways in protecting the rest of the respiratory tree when the daughter products are inhaled in very low dust atmospheres.

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Table 31.

Ratio of Maximum Local Dosage to Average Dosage in the Lung for Different Sized Particles

Diameter mu	Max. Dose / Avg. Dose	Region of Max. Dose
6	670	Quart. Br.
2	16	Quart. Br.
0.6	5	Resp. Br.
0.2	5	Resp. Br.
$D=16x10^{-6}$ [a]	8	Ter. Br.
D=0.03 [b]	320	Sec. Br.
Radon [c]	27	Trachea

[a]Diffusion coefficient for atmospheric condensation nuclei.

[b]Diffusion coefficient for positive ions.

[c]Dosage from radon and daughter products of radon molecules decaying in lung.

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11. DISTRIBUTION OF RADIOACTIVITY IN AIR CONTAINING RADON.

From the results given in Table 31, it is seen that the dosages imparted to the different regions of the respiratory tract depend on the nature of the carriers of the air-borne daughter products. The greatest local dosages for a given per cent deposition occur when the daughter products are inhaled as they were originally formed, that is, as positive ions. These dosages are to the upper regions of the respiratory tract. When the daughter products lose their mobility by becoming attached to the submicroscopic condensation nuclei normally present in the air, they are carried much farther into the respiratory tree before diffusing to the walls. Most of the alpha energy released by the daughter products when they are carried on the condensation nuclei is absorbed in the alveolar regions. Because the surface to volume ratios of these regions are very large, the alpha dosages produced by the deposited daughter products are the least in the respiratory tract.

11.1. Combination of Daughter Products with Atmospheric Nuclei.

About 90 per cent of the daughter products are positive ions at the end of the recoil which accompanies their formation (13). The fate of positive ions in the atmosphere has been studied extensively (52-56). These small ions disappear by recombination with negative small ions, by affixing to particulate matter in

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the air, and, if radioactive, by decaying. At equilibrium the rate of formation equals the rate of disappearance and we can write

$$\beta_1 n_1 = \alpha n_2 n_1 + \eta_{10} N_0 n_1 + \eta_{12} N_2 n_1 + \lambda_1 n_1$$

n_1 = concentration of + small ions

n_2 = concentration of - small ions

N_0 = concentration of uncharged condensation nuclei

N_2 = concentration of - charged condensation nuclei

λ_1 = decay constant for the radioactive ions

If we neglect the role of charge in the deposition of the daughter products we can write for their disappearance

$$\beta n = \eta N n + \lambda_1 n$$

n = concentration of daughter products existing as free molecules
η = combination coefficient for condensation nuclei
N = concentration of condensation nuclei

Thus we can calculate the ratio between daughter products which have and which have not combined with the atmospheric nuclei. Some published values of the concentrations of these nuclei are (40):

50/cc	for country air
5000/cc	for city air
100000/cc	for city air

η is of the order of 10^{-6} cm ^{3}/sec. (54)
λ = 38×10^{-4}, 4.3×10^{-4} and 5.9×10^{-4} sec.$^{-1}$ for RaA, RaB and RaC-C' respectively.

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For 50 nuclei/cc the fraction of uncombined daughter products will be

38/38.5 or 0.99 for RaA
(4.3/4.8) x 0.99 or 0.89 for RaB
(5.9/6.5) x 0.89 or 0.81 for RaC-C'

For 5000 nuclei/cc the fraction of uncombined daughter products will be

38/88 or 0.43 for RaA
(4.3/54.3) x 0.43 or 0.08 for RaB
(5.9/55.9) x 0.08 or 0.08 for RaC-C'

For 100000 nuclei/cc. a negligible number of the daughter product molecules remain uncombined.

The proportion of the uncombined atoms that become deposited on the walls of the enclosure containing the radon is a function of the diffusion coefficient of the daughter products, the circulation of the air in the enclosure, and the ratio of the surface area of the enclosure to the volume of gas within it.

11.2. Removal of Daughter Products by Surfaces.

We recall that turning a fan on with a low dust atmosphere in the chamber decreased the measured daughter product activity in the air by an order of magnitude (see Fig. 10). Stirring the air raised the concentrations of the daughter products next to the walls and increased the efficiency of the walls in removing them. We may consider what happens in the extreme case when stirring is so effective that the daughter product concentrations

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are always uniform throughout the chamber. From kinetic theory we calculate the number of collisions/cm ^{2}/sec by gas atoms against the walls of their container equal Nu/4 where N is the number of atoms/cm ^{3} and u is their velocity. The mean velocity of the daughter product atoms is about 1.5×10^4 cm /sec. Thus, the number hitting per second is 4000N and since there are approximately 10^5cm.^{2} of surface in the chamber, the number hitting the walls per second is 4×10^8N. Since the decay constant for RaA is 0.004 sec.$^{-1}$ and the volume of the chamber is 2×10^6cm.^{3}, we can write for the disappearance of the daughter products from the chamber

$$\frac{dN}{dt} = -0.004N - 4 \times 10^8 N$$

The ratio of the number that decay in the chamber to the number that are lost to the walls is $(4 \times 10^{-3})/(4 \times 10^8)$ or 10^{-11}. This result shows that in the extreme case considered, all the daughter products are deposited on the walls.

A fanning experiment was performed after creating a high dust load in the chamber with a NaCl aerosol. Fanning the air under these conditions decreased the daughter product concentration in the chamber by only 20 per cent. Apparently the daughter products were deposited on dust particles before getting to the walls. They remained in the chamber because, even with fanning, the dust particles were removed from the chamber relatively slowly.

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To obtain further information on the state of the airborne daughter products, cascade impactor samples were taken of the air in the chamber. Air drawn through the impactor is directed against four microscope slides in succession at increasing velocities and particles of successively smaller size are deposited on the slides. All the air is then drawn through a millipore filter which removes the remainder of the particulate matter in the air.

One sample was taken of air that had been in the chamber for eight days and thus had a fairly low dust load. After the sample was taken, a NaCl aerosol was inserted into the chamber. When radioactive equilibrium was attained, a second cascade impactor sample was taken. The results are given in Table 32.

Table 32.

Radioactivity Deposited on the Stages of the Cascade Impactor by Air Sampled from the Exposure Chamber

	Measured activity				
	Stage 1	Stage 2	Stage 3	Stage 4	Filter Paper
a) Low dust air	486	694	558	408	6819
b) High dust air	6	37	350	6850	47300
Stage calibration: Median size of deposited particles	13.1mu	5.4mu	2.85mu	1.9mu	-----

Volumes of samples (a) and (b) were approximately equal.

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We see that most of the activity was measured on the filter paper. This is to be expected since the slides do not remove many particles smaller than 1.9 mu. However, a relatively higher activity was measured on the first stages when the low dust air was sampled than when the air contained the added aerosol. Because the daughter products existed in a more mobile form in the air with the lower dust load they were deposited on the slides to a greater extent. These results point out that a given daughter product level is much more hazardous in a filtered than in an uncleaned atmosphere since a significant number of the daughter products suspended in filtered air are removed in the anterior regions of the respiratory tract where high dosages can be produced from the deposition of a very small percentage of the inhaled radioactivity.

12. SUMMARY AND CONCLUSIONS.

Well-type gamma counters of high efficiency were used to measure the radioactivity of the lungs removed from rats and dogs exposed to air containing known concentrations of radon and its daughter products. A two thousand liter exposure chamber was used to hold the radon atmosphere. The rats, all males weighing between 190 and 250 grams, were inserted directly into the chamber. The tracheae of the dogs were connected to the chamber by tracheal cannulae.

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Measurements were made of the retention of the daughter products breathed from the chamber by a human subject. A special technique was devised for sampling the expired air of the subject with a millipore filter.

12.1. Results and Conclusions from Experiments with the Rat.

1. The average gamma activity measured in the lungs of rats which had inhaled radon and its daughter products in atmospheric air that had not been cleaned was reduced 8.5 times in further rat experiments by filtering the air before inserting it into the exposure chamber and 60 times by fanning the air in addition to filtering it.

2. The gamma activity of the lungs was approximately proportional to the alpha or gamma activity of a millipore filter paper which had removed the daughter products from a small known quantity of chamber air.

3. The ratio of lung activity to filter paper activity was not significantly different for exposures using radon mixed with the following chamber atmospheres: (a) air inserted into the chamber from the out-of-doors a few hours before the animal exposure; (b) air confined in the chamber for several days before the exposure; (c) filtered air; (d) air to which cigarette smoke or a fine ZnO_2 dust had been added. However, the proportionality of lung activity to RaC' concentration was probably only coincidental

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for the lower dust loads. The RaC' concentration was measured in the chamber proper and independent experiments showed that at very low dust loads, the RaC' concentration in a dummy cage was much smaller than the concentration in the chamber.

4. The ratio of the activity in the lungs to activity on the filter paper was the same for rats inserted into the chamber in cages as for rats inserted into the chamber tied down to rat boards. The rats in the cages were usually quiet during the exposure. The rats tied down to the rat boards struggled to free themselves much of the time they were in the chamber.

5. Since the gamma activity of the lungs is proportional to the dosage imparted to the lungs by the radon daughter products, the experiments demonstrated that the filterable activity in the air may be used as an index of the dosage imparted by the radon daughter products to the lungs of rats breathing in air containing radon. This conclusion should also apply to the dosage imparted to the lungs of humans breathing in air containing radon. If measurements of the radon in the air are also available, the calculated dosage from radon in equilibrium with its daughter products in the lung may be added to the dosage obtained by the filter paper method. However, the dosage from inhaling the radon is generally negligible compared to the dosage from inhaling the daughter products.

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6. The average daughter product dosage to the rat's lungs, extrapolated to a rat breathing 10^{-11} c/l radon in equilibrium with its daughter products was 0.17 mrep/hr (3.3 mrem/hr). This was five times higher than the dosage measured in dogs and calculated for humans. The higher dosage to the rat should be taken into account in applying the results of chronic and acute exposures of rats to radon to an evaluation of the hazard to humans.

12.2. Results and Conclusions from Experiments with the Dog.

1. The total radioactivity measured in the lungs of dogs, connected to the radon exposure chamber through tracheal cannulae, did not depend very strongly on the respiratory minute volumes of the dogs.

2. The average daughter product dosage to the lungs of anesthetized dogs, extrapolated to a dog breathing 10^{-11} c/l radon in equilibrium with its daughter products was 0.041 mrep/hr. (0.82 mrem/hr).

3. The calculated dosage to the trachea from measurements of its radioactivity was ten times the average dosage to the lung.

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12.3. Results and Conclusions from Experimental and Theoretical Study of Human Exposure to Radon.

1. A human subject breathing from a chamber containing uncleaned atmospheric air mixed with radon retained 25 per cent of the inhaled daughter product activity. When breathing from an atmosphere of radon mixed with filtered air, he retained 75 per cent of the inhaled daughter product activity. His minute volume and tidal volume were about 10 liters and 1.3 liters, respectively.

2. The average dosage to the lungs produced by retaining 25 per cent of the inhaled RaA, RaB, and RaC-C' in equilibrium with 10^{-11} c/l radon is 0.033 mrep/hr (0.66 mrem/hr), assuming a lung weight of one kilogram. The average dosage to the lungs produced by the inhaled radon and the daughter products of the radon molecules decaying in the lungs is 0.0013 mrep/hr (0.026 mrem/hr).

3. Much larger dosages are calculated for local regions in the lung. The maximum dosage from inhaled daughter products carried on typical atmospheric nuclei (0.06 mu diameter) is imparted to the tertiary bronchioles in Findeisen's idealized model of the lung (see Table A.1, Appendix 2). This dosage is 6 mrem/hr. When the daughter products exist in a more mobile form, as in filtered atmospheres, the upper respiratory tract receives a much larger relative dosage than when the daughter

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products are carried on atmospheric nuclei.

The trachea gets the maximum dosage of 0.7 mrem/hr from the alpha particles emitted by the inhaled radon and the daughter products of the radon molecules decaying in the lung.

These values may be compared with the radiation tolerance dose for continuous exposure of human tissue of 1.8 mrem/hr used by the Subcommittee on Permissible Internal Dose of the National Committee on Radiation Protection to calculate tolerance levels for internal emitters. The level of 10^{-11} c/l radon in equilibrium with its daughter products has been recommended by the Subcommittee as the maximum permissible concentration level for continuous exposure.

12.4. Results and Conclusions from Sampling Studies.

1. A grab sampling procedure for determining the air-borne daughter product concentration, using an AA millipore filter to remove the daughter products from the air, was simple and reproducible.

2. The results of slow sampling, in which a liter of air was drawn through the millipore filter in ten minutes with a water-displacement aspirator, indicated that this method could be developed to give an indication of the mobility of the daughter products in the air, when compared to the results of a grab sample of an equal volume of air.

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3. Calculations show that the dosage to the lungs from inhaling the daughter products over an extended period of time is very nearly proportional to the activity of a grab air sample after the activity has decayed for about an hour, all other conditions being equal. It is not necessary to obtain the relative proportions of RaA, RaB, and RaC-C' in the inspired air.

12.5. Recommendations for Future Research.

Future animal experiments should be planned to study the pattern of deposition in the lungs as well as the total deposition. Activity measurements should be made on those larger passageways which can be dissected out of the lung. The longer-lived thoron daughter products could be used to obtain radioautographs of the deposited activity. They would be especially useful in studying deposition at the branch points in the lung.

The question of initial deposition versus retention over several hours should also be studied. The apparatus designed to sample the expired air of human subjects with millipore paper could be used to measure the initial deposition of the daughter products in experimental animals. This could be compared with the radioactivity of the lung after it had inhaled the daughter products for a long period of time. The biological half-life of the daughter products could also be obtained by measuring the time for equilibrium between deposition and decay of

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the daughter products in the lungs of experimental animals, e.g. dogs. The measurement could be made by placing a GM tube against the chest of the animal. A comparison of the measured time to reach radioactive equilibrium with the theoretical time to reach equilibrium if the daughter products were not removed from the lungs would give the biological half-life.

One could profitably accompany such studies with additional investigations of the general nature and efficiency of daughter product deposition on particles and surfaces. The role of charge in affecting deposition of the daughter products should be studied since the ratio of charged to uncharged daughter atoms depends on the total ionization in the air, and therefore on the radon concentration. Measurements of deposition in moist tubes, possibly in vitro experiments on human bronchial tubes, would yield particularly useful information, since the dosages to the larger bronchial tubes may be the critical dosages in radon exposure.

All these studies should be made using both daughter products which are carried on normal atmospheric dust and air-borne daughters which are suspended in cleaned air. In the first case, the daughter products should be deposited largely in the alveolar regions, while in the latter case, because of their higher mobility, they should be deposited to a considerable extent in the upper regions of the respiratory tract.

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In the final analysis one must resort to clinical experience to definitely establish hazardous radon concentrations in the air. It seems advisable, on the basis of the calculations which have been presented, to pay special attention to the larger bronchial passageways for evidence of chronic irritation from radon and radon daughter product exposure.

The daughter products were investigated primarily in connection with their hazard in uranium mining. However, the investigation has demonstrated that they also provide a means of tagging aerosols. It is obvious that used as tracers, they can be an extremely useful tool in the investigation of problems in pulmonary physiology.

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APPENDIX 1.

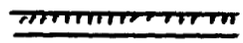
DERIVATION OF EQUATION FOR DETERMINATION OF ALPHA PARTICLE SELF-ABSORPTION IN MILLIPORE AIR SAMPLES

The equation for the alpha particles emitted from the source of Fig. A-1 has been derived by Finney and Evans (57) as

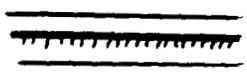
$$n_a = \frac{Nut}{4}\left[\frac{2(R-a)}{R} - t\right]$$

n_a = alpha particles emitted/cm^2
N = alpha disintegrations/cc
R = mean range of alpha particles (air-cm)
uR = range in source (cm)
ut = thickness of source (cm)
t = thickness of source (air-cm)
a_i = thickness of absorber (cm)
a_i/u_i = thickness of absorber (air-cm)

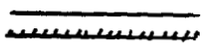
We apply this equation to the three measurements of Section 5.1.



Measurement 1. Normal measurement.



Measurement 2. Activity measured after sample covered with unused millipore filter.



Measurement 3. Activity measured when millipore placed in counter upside down.

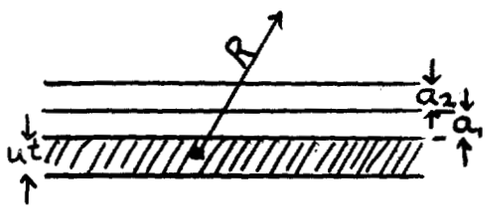


Fig. A-1. Alpha particle source and abosrbers.

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Measurement 1. $n_1 = \frac{Nut}{4}\left[\frac{2R-t}{R}\right]$

Measurement 2. $n_2 = \frac{Nut}{4}\left[\frac{2(R-a)-t}{R}\right]$

Measurement 3. $n_3 = \frac{Nut}{4}\left[\frac{2(R-a+t)-t}{R}\right]$

$$\text{Fraction transmitted} = \frac{n_1}{\frac{Nut}{2}} = \frac{2R-t}{2R}$$

$$n_3 - n_2 = 2\,\frac{kt}{R} \qquad K = \frac{Nut}{4}$$

$$t = \left(\frac{n_3 - n_2}{2n_1}\right)(2R-t)$$

$$t = \frac{2Ry}{1+y} \quad \text{where} \quad y = \frac{n_3 - n_2}{2n_1}$$

$$\text{Fraction transmitted} = \frac{2R - \frac{2Ry}{1+y}}{2R}$$

$$= \frac{1}{1+y}$$

$$= \frac{2n_1}{2n_1 + n_3 - n_2}$$

In this derivation it has been assumed that the radioactivity is deposited uniformly in the filter paper. Since the self-absorption is very small, the neglect of the true pattern of deposition will only affect the calculation of the transmission through the paper slightly. If the concentration of radioactivity decreases from the surface of the filter paper down, the assumption of uniform deposition will underestimate the transmission through the paper.

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APPENDIX 2.

CALCULATION OF DOSAGES TO THE RESPIRATORY TRACT FROM DEPOSITION OF RADON DAUGHTER PRODUCTS

The equation giving the fraction of particles removed by diffusion to the walls from an air stream flowing through a tube is (52):

$$f_\alpha = 1-0.819e^{-7.314h}-0.0975e^{-44.6h}-0.0325e^{-114h}-\ldots \qquad h>0.0156$$

$$f_\alpha = 4.07h^{2/3}-2.4h-0.446h^{4/3} \qquad h<0.0156$$

$h = \frac{D\tau}{2r^2}$ D = diffusion coefficient in cm^2/sec

$\tau = \frac{\text{volume of tube (cc)}}{\text{flow rate through tube (cc/sec)}}$

r = radius of tube (cm)

Laminar flow is assumed. A plot of f_α vs. h is given in Fig. A.2.

To obtain the deposition in a given tube of the respiratory tract during one respiratory cycle:

1. Calculate the deposition from inhaled air which flows completely through the tube.
2. Calculate the deposition from air which comes to rest in the tube.

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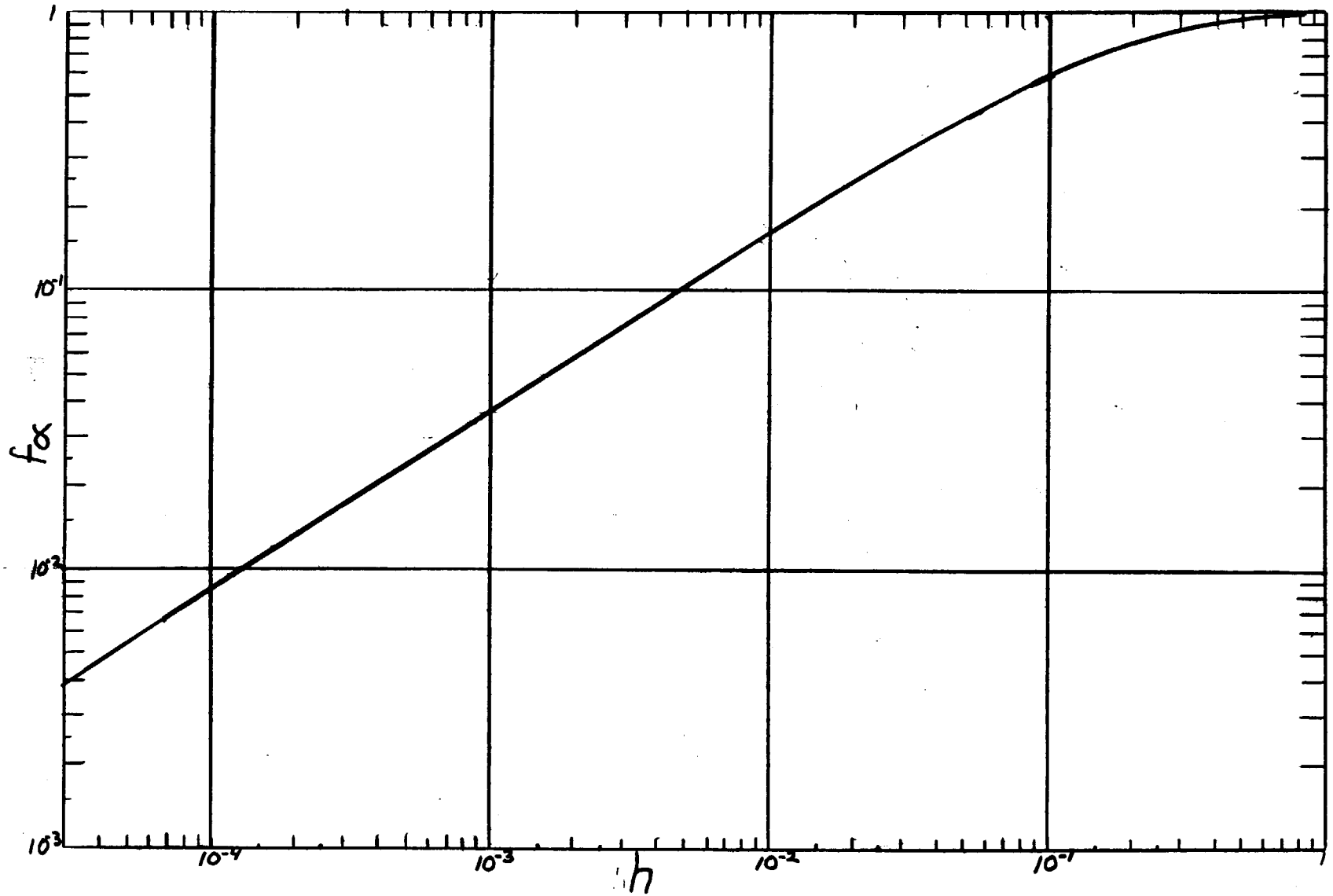

Fig. A.2. Plot of equation of f_α vs. h

If n(t) is the fraction of incoming particles which are still in the air stream after traveling through the tube for a given time t, then the fraction of incoming particles which still remain in the air stream on being exhaled back out of the tube is

$$\frac{n(\tau_p)\int_0^{t'} n(t)^2\,dt}{t'}$$

where t' is the time in which the first air entering the tube comes to rest and τ_p is the length of the pause after inhalation. The fraction transmitted can also be written

$$\frac{n(h_p)\int_0^{h'} n(h)^2\,dh}{h}$$

As an approximation, it can be assumed in most of the calculations that the mean concentration in the tube for air coming to rest in the tube is the average of the concentration at the beginning and end of the tube. The deposition must be obtained by integration only when the deposition is extreme.

3. Calculate the deposition from exhaled air which flows completely through the tube.

The total deposition is the sum of (1), (2) and (3).

A sample calculation of the deposition for a given set of conditions is illustrated in Table A.2. The schematic representation of the respiratory tract given in Table A.1 was used in the calculations.

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Table A. 1.

Schematic Representation of the Respiratory Tract
from H. D. Landahl, Bull. Math. Biophys. 12, 43 (1950)

Region α	Number	Diam. cm.	Cross section area	Lgth.	Volume	Σ Vol.	Passage Time 300cc/s	cc irradiated by α of Range 66 mu [a]	50 mu [b]
1. Mouth	1	2	3.14	7	22	22	0.073	0.292	0.219
2. Pharynx	1	3	7.07	3	21.2	43.2	0.071	0.187	0.142
3. Trachea	1	1.6	2.01	12	24.1	67.3	0.083	0.396	0.302
4. Prim. Bronchi	2	1	0.79	6	9.45	76.8	0.032	0.246	0.188
5. Sec. B.	12	0.4	0.13	3	4.53	81.3	0.015	0.300	0.228
6. Ter. B.	100	0.2	0.03	1.5	4.71	86.0	0.016	0.64	0.473
7. Quart. B.	770	0.15	0.02	0.5	6.81	92.8	0.023	1.25	0.940
8. Terminal B.	6×10^4	0.06	2.83×10^{-3}	0.3	51	143.8	0.17	24.8	18.4
9. Resp. B.	1.5×10^5	0.04	1.26×10^{-3}	0.15	28.4	172.2	0.094	21.7	18.7
10. Alv. Ducts I	3×10^6	0.03	7.04×10^{-4}	0.05	106	278.2	0.353	114	96.5
11. Alv. Ducts II	4×10^7	0.025	4.9×10^{-4}	0.03	588	866.2	1.96	788	660
12. Alv. Sacs	10^8	0.033		0.033					

[a] For 7.58 Mev., average energy of alpha particles from inhaled daughters.

[b] For 6.39 Mev., average energy of alpha particles from radon and its daughters in equilibrium.

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Table A. 2.

Sample Calculation of Deposition of Particles in Respiratory Tract by Diffusion

Tidal vol. = 450cc. Air Flow = 300cc/sec. Pause after inhalation = 0.5m.
Initial particle conc.=100/cc $D = 16 \times 10^{-6}$ cm ^{2}/sec.

c	h	Fraction retained	V_{in}	V_{out}	C_{in}	C_{out}	Mean conc. at end of insp.	Fraction retained during pause	Conc. at end of pause	$(C_{in}-C_{out} \times V_{out})$	$(C_{in}-C_p) \times V\alpha$	No. retained during expir.	Total No. Retained
1	58.3×10^{-8}	2.8×10^{-4}	450	428	100	100	100	0.001	100	12			12
2	24.5×10^{-8}	1.5×10^{-4}	428	407	100	100	100	0.0006	100	6			6
3	104×10^{-8}	4×10^{-4}	407	383	100	100	100	0.0015	100	15			15
4	102×10^{-8}	4×10^{-4}	383	373	100	100	100	0.0026	100	15			15
5	3×10^{-6}	8×10^{-4}	373	369	100	100	100	0.01	99	29	4.5		34
6	12.8×10^{-6}	0.002	369	364	100	99.8	99.9	0.02	98	73	9.4	22	104
7	32.2×10^{-6}	0.004	364	357	99.8	99.5	99.65	0.03	96	107	27.3	61	198
8	15.1×10^{-4}	0.05	357	306	99.5	94.5	97	0.10	87	1531	610	759	2900
9	19.8×10^{-4}	0.06	306	278	94.5	88.5	91.5	0.16	76.5	1530	496	679	2705
10	125×10^{-4}	0.19	278	172	88.5	71.6	80	0.23	61.5	2900	2870	1582	7352
11	294×10^{-4}	0.31	172	0	71.6	Retention by integration= 56%							6900
12												20238	
												Total in % ret.	45000 45%

C_{in} = particles/cc in air flowing into tube
C_{out} = particles/cc in air flowing out of tube
C_p = particles/cc in air in tube at end of pause
V_{in} = volume of air flowing into tube
V_{out} = volume of air flowing out of tube

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Table A. 2(Continued)

Removal on Expiration

Concentration of air leaving region[a]												Region air started from[b]	Vol. of air(cc)
6		7		8		9		10		11			
T	D	T	D	T	D	T	D	T	D	T	D		
				85								8	51
			.3		2								
				70.5		74						9	28
	0.1		.3		3.5		2.5						
				50		53		56.3				10	106
	.1		.2		3		3.3		5.2				
				22.8		24		25.5		31.5		11	172
	.05		.1		1.2		1.5		6				

Total No. Deposited = Conc. x Vol.

	15	102		
3	8	99	71	
10	21	318	350	550
8.5	17	240	258	1032
22	61	759	679	1582

[a] T = particles/cc in air leaving region.
D = particles/cc deposited.

[b] Removal in starting region = 1/2 removed calculated for air passing through entire tube.

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