CONTRIBUTION OF RADON IN NATURAL GAS TO THE DOSE FROM AIRBORNE RADON-DAUGHTERS IN HOMES*

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Abstract

Data have been obtained on the radon concentration in natural gas supplied to several metropolitan areas in the United States. The average value of 20 pCi/l was selected to estimate the contribution of this source of natural radioactivity to doses from radon-daughters received by individuals in homes. Radon-daughter concentrations in the home atmosphere were calculated by use of computer programs for an 8000 ft³ house in which 27 ft³ of gas per day was used for cooking in an unvented kitchen range. The total estimated dose to the bronchial epithelium included contributions from radon plus daughters in the outside ventilation air, each of which was assumed to be present at a concentration of 0.13 pCi/l, and from the radon plus daughters in the natural gas. The latter contribution averaged approximately 3% of the total dose. There was a 3.5% decrease in the estimated total dose when the air change rate increased from 0.25 to 2.0 per hour. We conclude that radon and radon-daughters entering the home with natural gas produce a negligible fraction of the total dose to the respiratory system of home occupants from airborne radon-daughters.

INTRODUCTION

The presence of radioactivity in natural gas was noted early in this century (Satterly, *et al.*, 1904), and measurements of the radon content of gas were first reported in 1919 (Satterly, *et al.*, 1918-1919). Since that time, many determinations of the radon content of natural gas at the wellhead have been reported, but little information has become available on the concentration of this radioactive rare gas in the natural gas entering homes. Also, little effort has been devoted to evaluation of doses that gas users receive from this natural source of radioactivity. We devised a three-part program to supply the missing information. The first part consisted of gathering data on the radon content of natural gas being supplied to several metropolitan areas in the United States. The second part involved the calculation of radon-daughter concentrations in homes produced by use of natural gas in an unvented home appliance. The last phase combined data from the first two parts of the program with the average of published estimates of the dose conversion factor for radon-daughters deposited in the human respiratory system to provide dose estimates for exposure to this source of natural radioactivity. To put these doses in perspective, we made a comparison with the dose received from radon-daughters produced by decay of radon in the atmosphere. A literature survey (Barton, 1971) pointed to the need for this investigation; a preliminary report has been published (Barton, *et al.*, 1973a).

RADON AND RADON-DAUGHTER CONCENTRATIONS IN NATURAL GAS AT POINTS OF USE

Several possible methods of obtaining data on the radon content of natural gas points of use were considered; sampling of a large fraction of the gas supplied to several large metropolitan areas was selected. Table 1 shows the organization of this part of the program which extended over approximately 1 year in search for possible seasonal variations. Table 2 summarizes the results obtained, while individual values (or monthly averages where more than one sample was analyzed) are plotted in Figure 1. In addition to monthly pipeline samples in the New York area, daily samples were analyzed at the U. S. Atomic Energy Commission's New York Health and Safety Laboratory. Monthly averages of these data are included in Table 2. The average value for all sampling locations is 17 pCi/liter, and we selected the rounded figure of 20 pCi/liter for dose calculations.

No data are available on radon-daughter concentrations in pipeline gas. Two efforts to detect daughter products in New York City gas (Breslin, 1972) and in Colorado (Schiager, 1973) were unsuccessful. The New

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York City test was handicapped by the low radon concentration in the gas (~1 pCi/liter). The radon concentration in the gas used in the Colorado test was higher, and a minimum detectable level of 0.06 working levels at the 95% confidence limit was reported. Since one working level is defined as the concentration of daughters in equilibrium with 100 pCi/liter of radon (or as any combination of short-lived daughters that will result in the ultimate emission of 1.3×10^5 MeV of alpha energy per liter of air), the above detectable level corresponds to the daughters in equilibrium with 6 pCi/liter of radon.

It was also reported (Breslin, 1972) that the particle count in New York City pipeline gas was quite low. This would partially account for the failure to detect daughters, since they are charged ions when formed. Unless there are gasborne particles to which they can become attached, they would be expected to migrate quickly to the pipeline wall.

RADON AND RADON-DAUGHTER CONCENTRATIONS IN VENTILATION AIR

Results of numerous studies of radon concentration in the atmosphere have been reported in the literature. They are summarized, in part, in literature surveys (Barton, 1971 and United Nations, 1972). The ^{222}Rn concentration in air ranges from approximately 0.001 to 1.0 pCi/liter, while the ^{220}Rn (thoron) concentration is approximately a factor of 100 lower. We have neglected the thoron content of the air in our dose calculations; it is not present in natural gas at points of use because of its short half-life (54.4 s). Determinations of the concentration of individual radon-daughters in air have been made much less frequently than radon measurements, and data on simultaneous measurements of radon and its daughters are relatively scarce.

For our average or representative radon concentration in air, we have averaged the results of a compilation of published data on measurements in the United States (Lowder, *et al.*, 1971). The resulting value (0.13 pCi/liter) is below the middle of the range mentioned above, but, if values as low as 0.001 to 0.003 pCi/liter observed in coastal areas and islands (Blifford, *et al.*, 1956) were included in the compilation, the average would be even lower than 0.13 pCi/liter.

Because of the above-mentioned scarcity of data on radon-daughter concentrations relative to radon in the atmosphere, we have adopted the reference radioactive atmosphere used by Altshuler, *et al.*, (1964) which was used earlier by the Public Health Service (Holaday, *et al.*, 1957) in recommending the working level. Although this ratio was based primarily on measurements made in mine atmospheres, it appears to be reasonably consistent with the scarce data on the atmospheric radon-daughter ratio; that is, both higher and lower values have been observed for the different daughters.

EXPOSURE MODEL

The exposure situations considered in this study assume, as in earlier studies of doses from tritium in gas (Barton, *et al.*, 1973b), that 0.765 m^3 of gas is consumed per day in an unvented kitchen range located in 226.6-m^3 (92.9 m^2 floor area) house. We further assume that the gas combustion products are uniformly dispersed in the home atmosphere. The above-mentioned value for daily range consumption is an average value for gas usage in this appliance in the United States (Segeler, 1966).

In the earlier studies, we assumed an average air turnover or ventilation rate of one per hour. In this investigation, our calculations covered air change rates of 0.25 to 2.0 per hour because we lacked definitive information on the average air change rate in houses having gas ranges.

As was mentioned in the section on radon and its daughter concentrations at points of use, we do not know how much, if any, radon-daughters are in the natural gas when it enters homes. Because of this lack of data, we calculated doses for the limiting case in which the daughters are in equilibrium with the radon on leaving the pipeline, and compared the results with those for gas containing no daughters. We also estimated doses, with and without the assumed presence of radon and its daughters, in the ventilation air. The different cases for which we estimated doses are described below.

Case 1. Natural gas containing radon at a concentration of 20 pCi/liter, but with no daughter activity, is used. This provides the lower dose limit. Ventilation air is assumed to contain no radon or radon-daughters. Although this case is not a practical situation, it permits calculation of the dose effect of radionuclides resulting from radon in the gas without the complication of ventilation air activity.

Case 2. The exposure conditions are identical to those for case 1 — except that the natural gas is assumed to also contain radon-daughters (RaA, RaB, and RaC) at their equilibrium concentrations (20 pCi/liter). This case provides an upper limit value for the dose from natural radioactivity in the natural gas for the specific conditions considered.

Case 3. The exposure conditions are identical to those for Case 1 — except that the ventilation air is assumed to contain radon and its daughter radionuclides (RaA, RaB, and RaC) each at a concentration of 0.13 pCi/liter. This case provides the lowest dose limit for the effect of radon in natural gas when the ventilation air also contains radionuclides.

Case 4. The exposure conditions are identical to those for Case 1 with two exceptions: (1) the natural gas is assumed to also contain radon-daughters at their equilibrium concentrations; and (2) the ventilation air is assumed to contain radon and its daughter radionuclides each at a concentration of 0.13 pCi/liter. This case provides an upper dose limit for the conditions under which Case 3 gives the lower limit.

Case 5. The exposure conditions are identical to those in case 3 — except that the daughters are assumed to have the distribution of the reference atmosphere of Altshuler, *et al.*, (1964) rather than being in equilibrium with radon. Since it appears probable that little, if any, daughter activity is in the gas when it enters homes, and that the daughter activity in the atmosphere is not generally in equilibrium with radon, this case is the most realistic of the five cases considered in regard to assumptions.

CALCULATION OF RADON-DAUGHTER CONCENTRATIONS IN HOMES

Radon decays according to the following scheme:

$$^{222}\text{Rn} \xrightarrow[3.823\ \text{days}]{5.49\ \text{MeV}\ \alpha} {}^{218}\text{Po(RaA)} \xrightarrow[3.05\ \text{min}]{6.00\ \text{MeV}\ \alpha} {}^{214}\text{Pb(RaB)} \xrightarrow[26.8\ \text{min}]{0.7\ \text{MeV}\ \beta} {}^{214}\text{Bi(RaC)}$$

$$\xrightarrow[19.7\ \text{min}]{3.3\ \text{MeV}\ \beta} {}^{214}\text{Po(RaC')} \xrightarrow[164\ \mu\text{s}]{7.687\ \text{MeV}\ \alpha} {}^{210}\text{Pb(RaD)} \xrightarrow[21\ \text{year}]{0.02\ \text{MeV}\ \beta} {}^{210}\text{Bi(RaE)}$$

Only the first three daughter products (RaA, RaB, RaC) need to be considered in making dose calculations, but the energy contribution of RaC', an alpha emitter, is attributed to RaC, because it has such a short half-life that its decay immediately follows that of its parent radionuclide (^{214}Bi RaC). Although RaB is a β-emitter and does not contribute significantly to the total dose, our dose calculations take into consideration its decay to RaC'. The very long half-life of RaD, in addition to its soft beta emissions, makes it unimportant from the radiation dose standpoint.

When the range is turned on, radon in the gas combustion products is dispersed in the home and the concentration of radon-daughters from this activity source begins to build up. At the same time, the daughters are removed by radioactive decay and by ventilation. A computer program was written to handle the calculation of radon-daughter concentrations in this dynamic situation. Mathematical analysis has demonstrated that the average 24-hour concentration of radionuclides is not affected by range-use schedule. In other words, it makes no difference whether the gas is used in three 1-hour periods during the day or all in one 3-hour period during the day. The important variables are the volume of gas used, the radon concentration in the gas, and the home ventilation rate. Our computer model assumes that the total average daily consumption of gas is burned in the range at a constant rate in one hour. The program calculates the number of atoms of radon, and each of the three daughters, for each second of a 24-hour period, and the results at each 60-s interval are included in the computer printout, as well as the cumulative average number of atoms of each species for a given radon input and ventilation rate.

For the even more complex situation, in which radon-daughters are present in the natural gas and/or in the ventilation air, another computer program, while less exact mathematically than the above-mentioned program, was developed that gives results which agree with the other program within 1 or 2%.

The average 24-hour values for the number of atoms of each radon-daughter are converted to concentrations by assuming that the gas combustion products are dispersed uniformly in the 226.6-m^3 house. They are then converted to working levels, as defined previously, by use of the known decay constants and decay energies for the individual daughters.

DOSIMETRY

The calculated concentrations of radon and radon-daughters in the home atmosphere were converted to estimates of radiation dose by using a dose conversion factor selected on the basis of a literature survey. Because of the complexity and specialization of radon-daughter dosimetry in the respiratory system, we concluded that our current interests do not justify independent development of the necessary factor. Data from the survey are summarized in Table 3. The dose conversion factors (rads/year) in the table are for an assumed continuous inhalation of radon-daughters at a concentration of one working level (WL). Discarding the highest and lowest factors in Table 3, the average value of the five remaining factors is 85 rads/year. Walsh (1970) reviewed the literature regarding radiation dose to the respiratory tract of uranium miners from inhalation of radon-daughters, and concluded that the average dose to the bronchial epithelium of the tracheobronchial tree from an exposure to radon-daughters at 1 WL for one year is not larger than 50 to 100 rads. and that the dose to the basal cells may be less than 50 rads. He pointed out, however, that localization of activity (e.g., at bifurcations) could produce much higher doses. In a report from the epidemiological study of United States uranium miners, Lundin, *et al.*, (1971) concluded that one year of continuous exposure to radon-daughters at 1 WL is equivalent to approximately 103 rads averaged over the tracheobronchial epithelium. Evans (1967) has concluded from the work of Altshuler, *et al.*, (1964) and Jacobi (1964) that the dose conversion factor for inhalation of radon-daughters ranges from approximately 25 to 160 rads per year of continuous exposure at 1 WL. The dose conversion factor selected for use in this report is 100 rads to the bronchial epithelium per year of continuous exposure to radon-daughters at a concentration of 1 WL. The basal cells of the bronchial epithelium are assumed to be the critical tissue. An estimate of the corresponding dose to the total lung mass is given by Holleman (1968); based on uniform deposition of the alpha energy in a 1,000-g lung, the organ dose is approximately an order of magnitude less than the dose estimated for the bronchial epithelium.

Additional considerations in our treatment of the problem should be noted. These considerations, which may influence the reader's interpretation of the dose estimates presented, are:

(1) Use of the WL concept implies that the relative concentrations of RaA, RaB, and RaC in the inhaled air are not of major importance for dose calculation — in spite of the difference in the alpha decay energy of RaA and that of RaC' to which all three daughters decay. However, there are differing opinions on this point. For example, Lundin, *et al.*,(1971) state that the relative concentrations of RaA, RaB, and RaC are not of major importance for dose calculations; while Harley, *et al.*,(1972) state that the alpha dose for 1 WL may be widely different depending on the ratios of the radon-daughters.

(2) The dose contributions from inhaled radon, and from the decay of radon, or its daughters, absorbed in tissue have been ignored. Work reported by Holleman (1968) indicates that the absorbed radon and radon-daughter dose component adds only a small (0.5%) dose. For the exposure conditions specified in this report, the radiation dose is primarily due to inhaled radon-daughters (Shapiro, 1954).

(3) We adopted a quality factor (QF) of 10 for alpha particles in converting our dose estimates from rads to rems — following the current recommendations of the International Commission on Radiological Protection (1966). Some investigators adopt other values for QF. For example, Lundin, *et al.*, (1971) selected a QF of 3; however, most investigators express their results only in rads because of the lack of agreement on the appropriate QF for alpha particles.

RESULTS AND DISCUSSION

Estimated doses to the bronchial epithelium as a function of ventilation rate for Cases 1 and 2 are shown in Figure 2, while similar data for Cases 3 and 5 are displayed in Figure 3. The data for Case 4 are too close to those shown for Case 3 to make it practical to include them in the graph.

It is quite clear from Figure 2 that doses from radon, or radon plus daughters, introduced in gas vary quite markedly with ventilation rate. The assumed presence of daughters in the entering gas does increase the dose appreciably, but comparison of Figures 2 and 3 shows that the dose contribution from radionuclides in natural gas is small compared to that from ventilation air. The small variation in estimated dose with ventilation rate observed for Case 3 in Figure 3 is due to the radon-daughters from radon in natural gas. Since the daughters in ventilation air in this case are assumed to be in equilibrium with radon, changes in ventilation rate would not change the dose from this source, but daughters from radon in the gas will increase with decreasing ventilation rate. The relatively large effect of ventilation rate on total dose for Case 5 (Figure 3) is due to the assumed nonequilibrium daughter concentrations in this case. If the ventilation rate were zero, the daughter activities would soon be equal to the radon activity (0.13 pCi/liter).

Inspection of the data in Table 2 shows that, although the average of all the values is close to the 20-pCi/liter figure used in our dose calculations, the total range of radon concentrations at points of use is approximately 1 to 100. Doses in this range can be scaled directly from the values in Figure 2. Considering only the ventilation rate of one air change per hour, the above radon concentration range corresponds to doses varying from 0.75 to 75 millirems per year for Case and 1.4 to 141 millrems per year for Case 2. The maximum values are 6 to 11%, respectively, of the Case 3 and 4 values. Although the maximum doses are not insignificant, they are considered to be small as compared to probable variations in dose from radon and its daughters in air.

We have not previously mentioned a third source of airborne radioactivity in the home: radon and thoron from home construction materials. Reported measurements surveyed by Barton (1971) and others (United Nations, 1972) show that values vary widely with type of construction material and ventilation rate, so that it is difficult to arrive at an average or typical value. The mean of measurement in 324 dwelling places in Europe and the United States (United Nations, 1972, Table 13) is 0.52 pCi/liter, and values as high as 10 pCi/liter were reported in Europe and 4.8 pCi/liter (Lowder, *et al.*, 1971) in the United States. The mean of all measurements of outdoor radon concentrations made in connection with the indoor measurements quoted above is 0.084 pCi/liter. It appears, therefore, that the radon concentration in homes is likely to be much higher than the value assumed in our calculation (0.13 pCi/liter), which ignores the contribution of home construction materials, further reducing the significance of the contribution of natural gas activity to the total airborne natural radioactivity in homes. We conclude that other factors such as home construction material and ventilation rate are more important than the radon concentration in natural gas in determining the level of airborne natural radioactivity in homes.

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Table 1. Sampling and Analyses of Natural Gas.

Area	Pipeline Company	Organizations That Made Analyses	Organizations Bearing Analytical Costs
New York	Texas Eastern Transmission Company (TET)	AEC-New York Health and Safety Laboratory	AEC-New York Health and Safety Laboratory
New York	Transcontinental Gas Pipeline Corporation (TRANSCO)	AEC-New York Health and Safety Laboratory	AEC-New York Health and Safety Laboratory
Chicago	Natural Gas Pipeline Company of America (NGPL)	Argonne National Laboratory	Natural Gas Pipeline Company of America
Denver	Colorado Interstate Gas Company (CIG)	Colorado State University	Colorado Interstate Gas Company
Southwest and West Coast	El Paso Natural Gas Company (EPNG)	New Mexico Technical Research Foundation	El Paso Natural Gas Company

Table 2. Summary of Radon Measurements in Natural Gas Samples.

Area Served	Identification	Number of Samples	^{222}Rn Analysis (pCi/liter) Average	Range
Chicago	Amarillo	12	24.6	19.3 - 31.3
Chicago	Gulf Coast	12	3.2	2.3 - 4.4
New York City	TET	10	1.8	0.6 - 3.5
New York City	TRANSCO	8	1.4	0.6 - 1.6
New York City	TRANSCO-HASL	85	1.4	0.5 - 7.4
Denver	Wyoming	15(a)	5.8(a)	1.2 - 8.2
Denver	Kansas	8	91	15.3(b) - 118.8
Denver	Ft. Morgan (storage)	1	9.3	
El Paso	El Paso	2	17	
West Coast	Topock	--	19(c)	
West Coast	Blythe	--	9(c)	

(a) Includes examples taken at Ault and Aurora.

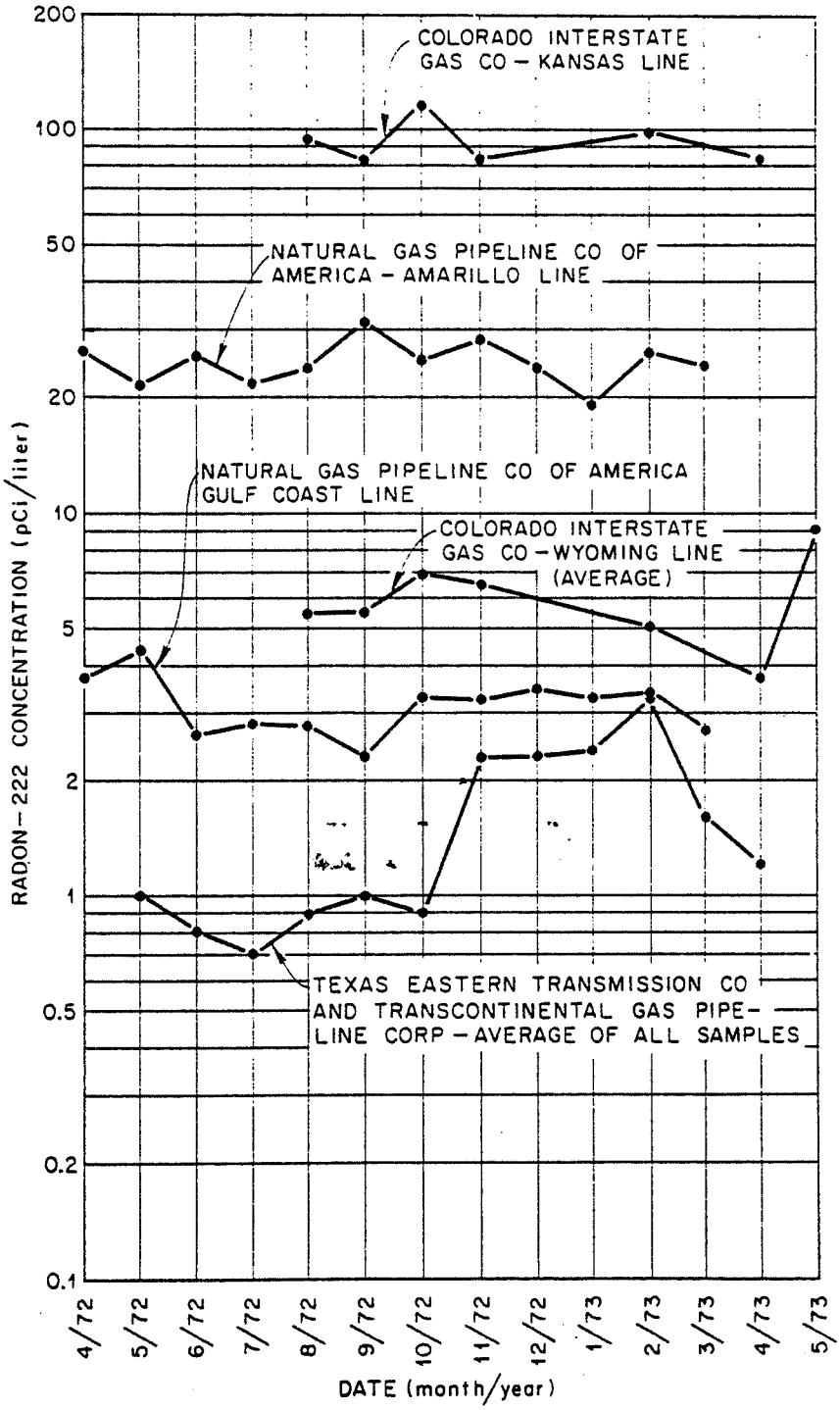
(b) Value excluded from average — sample taken during period of low gas usage.

(c) Values corrected for mixing and decay to the listed distribution point from analyses of samples taken at upstream sampling points. There would be further decrease of approximately 1 to 2 pCi/liter before the gas reached the Los Angeles market.

Table 3. Summary of Dose Conversion Factors for Radon and Radon-Daughters.

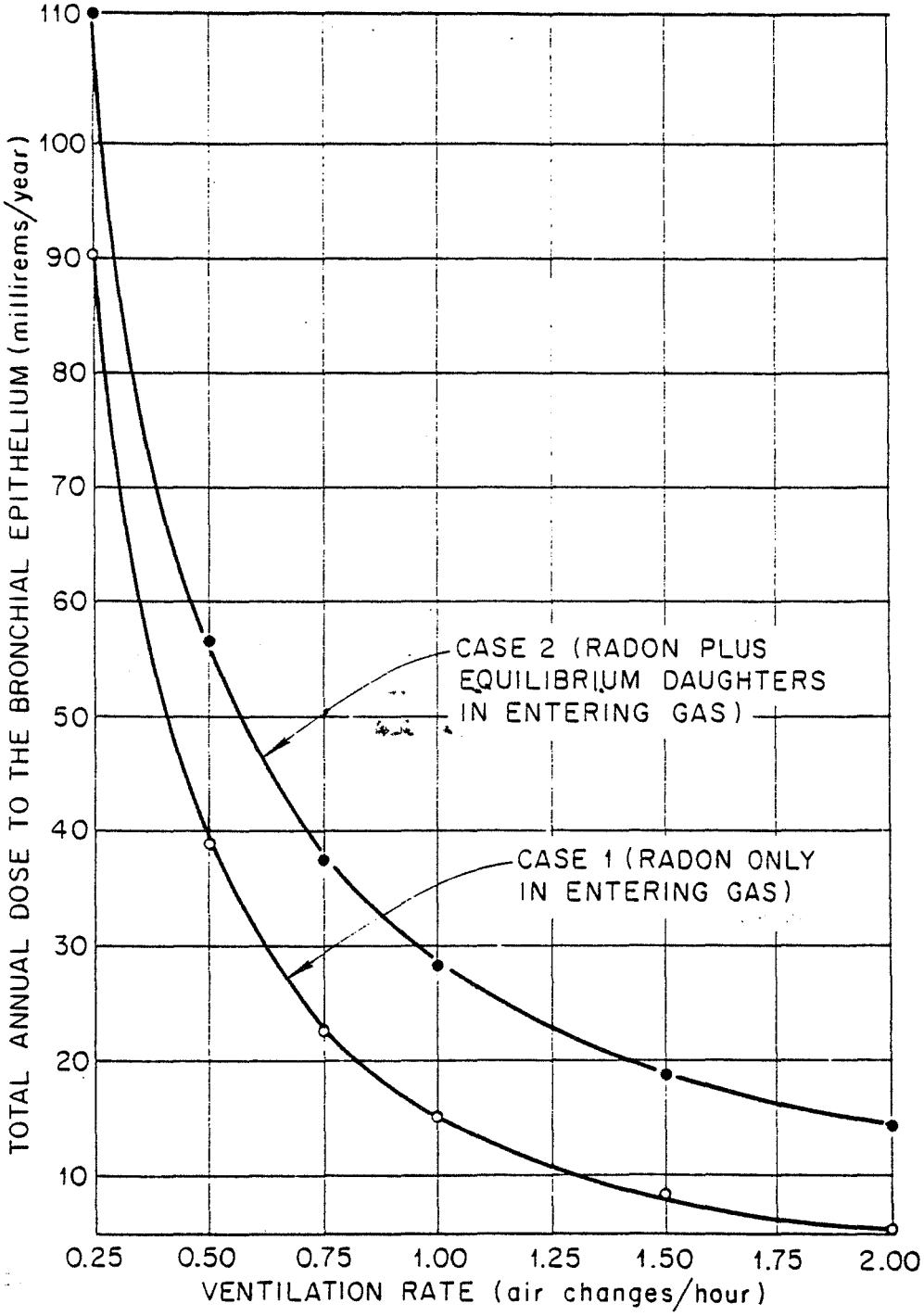
Isotopes Included	Critical Tissue	Calculated Dose (rads/year)(a)	Reference
Radon - daughters	Tertiary bronchioles	30	Shapiro (1954)
Radon - RaA	Main bronchi	20	Chamberlain, *et al.*,(1956)
Radon - daughters	Bronchial tissue	120	Holaday, *et al.*,(1956)
Radon-daughters	Segmented bronchi	150	Altshuler, *et al.*,(1957)
Radon-daughters	Secondary-quarternary bronchioles	150	Jacobi (1964)
Radon-daughters	Segmented bronchi	620	Hague, *et al.*,(1967)
Radon-daughters	Tertiary bronchioles	40	Holleman (1968)
Radon-daughters	Segmented bronchioles	12	Harley, *et al.*,(1972)

(a)The dose is calculated in rads per year for continuous exposure to radionuclides in a concentration equivalent to one "working level," defined as any combination of short-lived radon-daughters that will result in the ultimate emission of 1.3×10^5 MeV of alpha energy per liter of air.



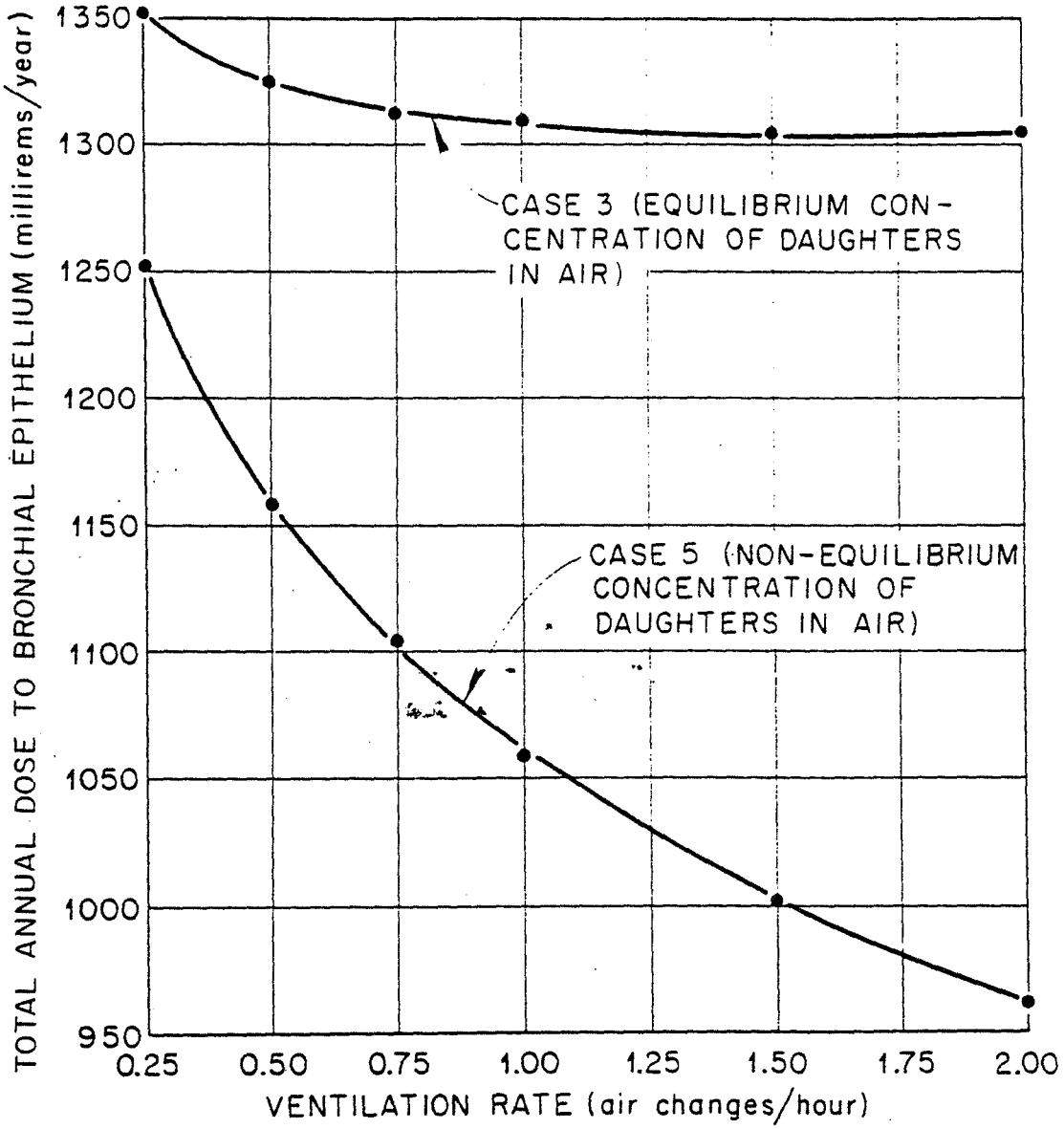
Observed Concentrations of Radon in Pipeline Gas Samples.

Figure 1.



Effect of Ventilation Rate on Dose from Radon Daughters Assuming No Activity in Ventilation Air.

Figure 2.



Effect of Ventilation Rate on Dose from Radon Daughters from Natural Gas and Air.

Figure 3.