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CONTRIBUTION OF RADON IN NATURAL GAS TO THE NATURAL RADIOACTIVITY DOSE IN HOMES

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OAK RIDGE NATIONAL LABORATORY
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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/liter 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-ft^3 house in which 27 ft^3 of gas per day is used for cooking in an unvented kitchen range. The total estimated dose to the bronchial epithelium included contributions from radon daughters in the ventilation air, each of which was assumed to be present at a concentration of 0.13 pCi/liter, and from 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 home dose to the respiratory system of home occupants from airborne radon daughters.

INTRODUCTION

A literature survey[1] revealed much data on the radon content of natural gas at the wellhead but almost no information on the concentration at points of usage. No estimates were found of the dose that is received by occupants of homes in which natural gas containing radon is used.

A three-part program described in this report was launched to supply the missing information which is of interest in connection with our dose estimations for uses of nuclearly stimulated gas. This investigation is continuing, but recent interest in an indication of the magnitude of this dose potential prompted estimates for the exposure situations considered in this report. A more thorough examination of this source of home radioactivity will be reported at the conclusion of our studies.

The first part of our program consists of obtaining data on the radon concentration of natural gas that is being supplied for consumption in several market areas. The second part involves calculation of the concentration of radon daughter products in the home atmosphere produced by use of natural gas in an unvented home appliance. The last phase combines data from the first two parts of the program with the best published estimates of the dose conversion factors for radon daughters deposited in the human respiratory system to provide dose estimates for exposure to this source of natural radioactivity and a comparison with the dose received from the normal atmospheric radon content.

The exposure situations considered in this study assume, as in earlier estimates of doses from tritium in Rulison gas,[2] that 27 ft^3 of gas is consumed per day in an unvented kitchen range located in an 8000-ft^3 house. However, in this investigation, our calculations covered air change rates of 0.25 to 2.0 per hour rather than the previously used rate of 1.0 per hour because of a lack of definitive information on the average air change rate in homes having gas ranges.

RADON CONCENTRATION IN NATURAL GAS AT POINTS OF USE

Establishment of a nationwide natural gas sampling and analysis program to determine an average radon concentration for the entire country appears to be impractical and unjustified. An alternative selected was the sampling of a large fraction of the gas supplied to several metropolitan areas. This part of the investigation was possible only through the cooperation of several gas transmission companies in providing monthly samples and, in some cases, paying for the analyses, as well as the help of several institutions in measuring the radon content of the samples. Table 1 shows the organization of this program. The results obtained to date in this continuing program are summarized in Table 2.

No data are available on the radon daughter concentration in pipeline gas. An attempt was made to detect radon daughters in New York City gas, but none were detected,[3] possibly because of the low (<1.0 pCi/liter) radon content of the gas. However, a mathematical evaluation of the possible consequence of daughters being present in equilibrium with radon compensated this lack of

Table 1. Sampling and Analyses of Natural Gas

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

data. For the dose calculations reported in this document, we have used the average of all the data in Table 2 as a guide in selecting a value of 20 pCi/liter for the concentration of radon in the gas. If the highest average value for a single sampling point (95 pCi/liter) is excluded, the average value for the remaining sampling locations is 10 pCi/liter.

RADON CONCENTRATION IN VENTILATION AIR

The previously mentioned literature survey[1] showed that reported values for the ^{222}Rn concentration in the atmosphere cover a range from 100 to 1000 pCi/m^3, while the ^{220}Rn (thoron) concentration is about 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 sec). A report[4] on measurements of radon in homes contains a tabulation of reported values for the radon concentration in the atmosphere in the United States. The average of these values is 0.13 pCi/liter or 130 pCi/m^3; we have adopted this figure as the concentration in ventilation air introduced into the home. We have neglected the contribution of radon and thoron from the home walls in making our estimates of doses in homes, because the concentrations of these gases and their daughters depend so markedly on the home construction material and on the ventilation rate that an average value for homes in this country cannot be postulated from the few studies that have been made in this area.

Table 2. Summary of Radon Measurements in Natural Gas Samples

Area Served	Sample Identification	Number of Samples	^{222}Rn Analysis (pCi/liter)	
			Average	Range
Chicago	Amarillo	8	25.6	21.9 - 31.3
Chicago	Gulf Coast	8	3.15	2.29 - 4.42
New York City	TET	6	1.7	0.55 - 3.8
New York City	TRANSCO	12[a]	1.2	0.5 - 3.5
Denver	Wyoming[b]	8	5.9	1.2[c] - 8.1
Denver	Kansas	4	95	15.3[c] - 118.8
El Paso	El Paso	2	17	
West Coast[d]	Topock	--	19	
West Coast[d]	Blythe	--	9	

[a]Includes seven samples of TRANSCO gas taken at the AEC-New York Health and Safety Laboratory.

[b]Includes samples taken at Ault and Aurora.

[c]Value excluded from average--sample taken during period of low gas usage.

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

CALCULATION OF RADON DAUGHTER CONCENTRATION IN HOMES

Radon decays according to the following scheme:

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

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

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 mother element (^{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 the α-emitter RaC'. The very long half-life of RaD, in addition to its soft beta activity, makes it unimportant from the radiation dose standpoint.

As stated in the introduction, the exposure situation that we are considering is the use of natural gas containing radon for cooking in a range located in a home with 8000-ft^3 volume and having an air change rate varying from 0.25 to 2.0 per hours. When the range is turned on, radon in the gas combustion products is dispersed in the home and the concentration of radon daughters 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-hr 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-hr periods during the day or in 1-hr followed by a 23-hr decay period. The important variables are the volume of gas used, the radon concentration in the gas, and the home ventilation rate. The program calculates the number of atoms of radon and each of the three daughters at 60-sec intervals and the cumulative average number of atoms of each for a given radon input and ventiliation 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-hr 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 8000-ft^3 house. They are then converted to "working levels" as defined in the following section by use of the known decay constants and decay energies for the individual daughters.

GENERAL CONSIDERATIONS AND FRAMEWORK OF DOSE ESTIMATION

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 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. A working level (WL) is defined as any combination of short-lived radon daughter products that will result in the ultimate emission of 1.3×10^5 MeV of alpha energy per liter of air. Discarding the highest factor and the lowest factor in Table 3, the average value of the five remaining factors is 85 rads/year. Walsh[13] 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.[14] 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 has concluded from the work of Altshuler et al.[8] and Jacobs[9] 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

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[5]
Radon + RaA	Main bronchi	20	Chamberlain and Dyson[6]
Radon + daughters	Bronchial tissue	120	Holaday et al.[7]
Radon daughters	Segmented bronchi	150	Altshuler et al.[8]
Radon daughters	Secondary-quarternary bronchioles	150	Jacobi[9]
Radon daughters	Segmented bronchi	620	Haque and Collinson[10]
Radon daughters	Tertiary bronchioles	40	Holleman[11]
Radon daughters	Segmented bronchioles	12	Harley and Pasternack[12]

[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.

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;[11] based on uniform deposition of the alpha energy in a 1000-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 the point. For example, Lundin et al.[14] state that the relative concentrations of RaA, RaB, and RaC are not of major importance for dose calculations, while Harley and Pasternack state in a recent publication[12] 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 decay of radon absorbed in tissue or of the daughters resulting from the absorbed radon have been ignored. Work reported by Holleman[11] indicates that the absorbed radon-radon daughter dose component adds only a small (0.5%) additional dose. For the exposure conditions specified in this report, the radiation dose is primarily due to inhaled radon daughters.[5] (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.[15] Some investigators adopt other values for QF, for example, Lundin et al.[14] 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.

ASSUMED RADON AND RADON DAUGHTER CONCENTRATIONS

Case 1. Natural gas containing radon at a concentration of 20 pCi/liter but with no daughter activity is used. Since no data are available on the concentration of radon daughters in natural gas entering homes, this provides the lower dose limit. The air turnover rate in the house varies from 0.25 to 2.0 per hour. 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.

RESULTS AND DISCUSSION

Results of our dose calculations are presented in Tables 4 through 7. These calculations cover a rather wide range of air change rates to compensate the lack of a representative value for the average rate for occupied homes having unvented kitchen ranges. The estimated total annual doses to the bronchial epithelium are compared in Table 8 for the two sets of closely related cases.

The data in Tables 4 and 5 show that there is a marked effect of air change rate on total dose in the absence of radon and radon daughters in the ventilation air. The ratio of Case 2 to Case 1 doses (4th column, Table 8) shows that the assumed presence of equilibrium concentrations of radon daughters in the natural gas used for cooking doubles the total dose at an air change rate between 1.0 and 1.5 per hour. However, for the more nearly realistic situation considered in Cases 3 and 4, with radon and radon daughters in the ventilation air, the assumed presence of daughters in the gas increases the total dose

Table 4. Estimated Doses to the Bronchial Epithelium from Radon Daughters as a Function of Air Turnover Rate (Case 1)

Number of Air Changes per Hour	Estimated Annual Dose to Bronchial Epithelium (millirem)			
	RaA	RaB	RaC	Total
0.25	11.2	47.6	31.4	90.2
0.5	5.57	20.9	12.4	38.9
0.75	3.68	12.3	6.62	22.6
1.0	2.71	8.18	4.06	15.0
1.5	1.76	4.43	1.90	8.09
2.0	1.28	2.76	1.04	5.08

Table 5. Estimated Dose to the Bronchial Epithelium from Radon Daughters as a Function of Air Turnover Rate (Case 2)

Number of Air Changes per Hour	Estimated Annual Dose to Bronchial Epithelium (millirem)			
	RaA	RaB	RaC	Total
0.25	11.5	56.6	41.7	110
0.50	5.8	28.7	21.2	56.7
0.75	3.9	19.3	14.2	37.4
1.00	2.9	14.5	10.7	28.1
1.50	2.0	9.7	7.1	18.8
2.00	1.5	7.3	5.3	14.1

Table 6. Estimated Dose to the Bronchial Epithelium from Radon Daughters as a Function of Air Turnover Rate (Case 3)

Number of Air Changes per Hour	Estimated Annual Dose to Bronchial Epithelium (millirem)			
	RaA	RaB	RaC	Total
0.25	142	701	510	1353
0.50	139	686	499	1324
0.75	138	680	497	1315
1.00	138	678	494	1310
1.50	137	675	494	1306
2.00	137	675	494	1306

Table 7. Estimated Dose to the Bronchial Epithelium from Radon Daughters as a Function of Air Turnover Rate (Case 4)

Number of Air Changes per Hour	Estimated Annual Dose to Bronchial Epithelium (millirem)			
	RaA	RaB	RaC	Total
0.25	142	709	520	1371
0.50	139	693	510	1342
0.75	138	685	504	1327
1.0	137	682	502	1321
1.5	136	680	499	1315
2.0	136	677	496	1309

by 1.3% or less. The slight discrepancy in the difference figures (columns 2 and 3, Table 8) can be attributed to the fact that computer calculations (carried to three significant figures) of the number of daughter atoms in the home as a function of time and air change rate were used to generate the four-digit doses in Tables 6 and 7.

The results presented in Tables 4 through 7 indicate that the dose contribution from radon and radon daughters which may be present in natural gas is small (approximately 2% of the total) when compared with similar estimates for the radon and radon daughters present in ventilation air. Reported concentrations of radon in the atmosphere vary by a factor of approximately 10.

Whether radon daughters are or are not present in natural gas appears to be of minor importance in light of the overwhelming dose contribution from radon and radon daughters present in ventilation air. We have not considered radon or thoron daughters from decay of these rare gases coming from building materials. From data in our literature survey,[1] it appears that the daughter concentrations in some homes may be as much as ten times the value (0.13 pCi/liter) for ambient air used in our dose calculations. This means that the contribution of radon daughters coming from the natural gas would be only about 0.1 or 0.2% of the total dose from airborne daughters in the home.

CONCLUSION

The potential dose from radon daughters produced by decay of radon introduced into a home with natural gas used in an unvented appliance is very small compared to the total dose from all airborne

Table 8. Comparison of Estimated Total Annual Doses to Bronchial Epithelium

Number of Air Changes per Hour	Difference Between Doses		Ratio of Doses	
	Case 2 - Case 1	Case 4 - Case 3	Case 2/Case 1	Case 4/Case 3
0.25	20	18	1.22	1.013
0.50	18	18	1.46	1.013
0.75	15	12	1.65	1.009
1.00	13	11	1.87	1.008
1.50	11	9	2.32	1.006
2.00	9	3	2.78	1.002

radon daughters, including those entering with the ventilation air and those resulting from decay of radon coming from home building materials.

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