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Measurement of Sources and Air Concentrations of Radon and Radon Daughters in Residential Buildings

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ABSTRACT

In the past several years measurements of radon and its daughters made in residential buildings in the U. S. indicate that in some situations the potential health hazard to the occupants of buildings from inhalation exposure can be significant. Measurement results from different geographical areas of the U. S. indicate that indoor radon and radon daughter concentrations can vary by as much as a factor of 100. In order to address this problem we will present methods and techniques necessary to determine the range and distribution of the annual average concentration of radon and daughters from which the annual mean radiation dose can be calculated. For diagnostic purposes and for the calculations necessary to determine the radiation dose to the lungs, we will describe additional measuring techniques for the sources of radon, air exchange rate, particle size of radon daughters and respiratory deposition.

INTRODUCTION

For the general public, the principal source of exposure to radiation is from inhalation of the short-lived daughters of radon. Most of this exposure occurs within their own homes, where people spend many hours each day, and where the radon levels are usually higher than outdoors. As residential ventilation is reduced through weatherization measures and new construction techniques, the concentration of radon, radon daughters, and other indoor pollutants can be expected to increase. This is of concern to the Department of Energy as it considers recommendations for methods of conserving fuel.

Concern about the health effects of radon and its daughters stems from the experience of the early uranium miners, some of whom were exposed to high levels of radon daughters in the course of their work. These miners showed an increased incidence of bronchial cancer many years after their exposure. Retrospective studies (Waggoner et al. 1963; Radford et al. 1984) have shown that the increase in risk was proportional to the accumulated exposure to radon daughers. These findings, together with advances in radiation biophysics, have made it possible to estimate the increasing cancer risk from the contribution of indoor radon and its daughters. The most recent report published on this subject was by the National Council on Radiation Protection and Measurements (NCRP 1984), which concludes with a recommendation that individuals should take action if their annual exposure to radon daughters exceeds 2 WLM (working level months).

Although several groups funded by the U. S. Government have conducted limited surveys for indoor radon and radon daughters, the accumulated data are not sufficient to estimate cancer risk country wide.

The purpose of this paper is to identify sources of radon and describe methods and monitoring techniques used to determine the radiation exposure of the general public to radon and its daughters inside buildings in different geographic locations. Measuring sources of radon may be useful in predicting what radon and radon daughter concentrations will appear in new construction and what to do in existing housing to minimize the impact of high concentrations.

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PHYSICAL CHARACTERISTICS OF RADON AND RADON DAUGHTERS AND THEIR RELATIVE RADIATION HAZARD

The noble gas ^{222}Rn is the immediate decay product of ^{226}Ra, a member of the decay chain of uranium, which is a naturally occurring trace element distributed in soil, rocks, and water. Ordinary soil and rocks contain about 1 pCi of $^{226}Ra.g^{-1}$. The decay chain of ^{222}Rn is listed in Table 1. Radon, because of its relatively long half-life compared to the time air is in the lungs, is not a significant source of exposure. However, its daughter products (^{218}Po, ^{214}Pb, ^{214}Bi, and ^{214}Po) have greater radiological significance, particularly the two alpha emitters, ^{218}Po and ^{214}Po. They have short half-lives and exist either as free atoms or ions or may attach to dust particles in air and may plate out on walls, furniture, and other surfaces. Under typical living or working conditions, most of the radon daughters attach to dust particles in air and about 20% to 50% (George and Breslin 1967) of those inhaled deposit on the epithelium lining of the respiratory system where they irradiate the basal cells.

A special quantity, the potential alpha energy concentration (abbreviated PAEC), has been devised to express the concentration of any mixture of short-lived radon daughters in air. The unit used for the potential alpha energy concentration is the working level (WL), defined as 1.28×10^5 MeV.L^{-1}, which is the summed energy of the alpha particles ultimately emitted by a mixture of 100 pCi each of ^{218}Po, ^{214}Pb, ^{214}Bi and ^{214}Po in 1 L of air. A term that is commonly used to describe the state of equilibrium between radon and radon daughters is the equilibrium ratio (ER). For example, in a typical house where the radon concentration in air is 1 pCi.L^{-1} and the working level from the four daughters is 0.005, the equilibrium ratio is

$$ER = \frac{0.005 \text{ WL} \times 100}{1 \text{ pCi.L}^{-1} \text{ radon}} = 0.5 \qquad (1)$$

In health physics, exposure to radiation by inhalation is defined as the product of concentration and time. One WLM is the occupational exposure to air containing 1 WL of radon daughters for a period of 173 h (a working month, WM). For the general public, exposure occurs for a period of 730 h each month, and the yearly exposure corresponding to PAEC = 1 WL is therefore (730 h.month^{-1})/(173 h.WM^{-1} x 12 months.yr^{-1} = 50.6 WLM.

Thoron and thoron daughters, like radon and radon daughters, are also present in small quantities in all rocks, soil, and building materials. Because of thoron's short half-life (55.6 s), a very small amount diffuses into the indoor environment and the buildup of thoron daughters is insignificant. However, there is limited evidence that the contribution of thoron daughters can be significant under some circumstances, one of which is likely to be a high thorium content in building materials. For this reason, some parallel measurements for thoron daughters must be made in different geographical areas to establish their contribution to the total radiation dose.

SOURCES OF RADON INDOORS

Radon, being the daughter product of ^{226}Ra, is formed whenever ^{226}Ra is present. Figure 1 shows the primary sources of radon inside buildings. In order of their usual contribution, they are the soil and geological substrate adjacent to the foundation, building materials, and tap water when it is supplied from water aquifers. Other sources that play minor roles are natural gas near the supply well, geothermal plant waste streams used domestically near the plant, and outdoor air in regions with extensive mineralization.

Soil as Radon Source

Most of the radon found indoors usually comes from the soil gas, which moves into the indoor environment by diffusion and convection. At a soil surface with natural cover, the radon flux ranges between 10-500 x 10^{-18} Ci cm^{-2} s^{-1}. The world average was found to be 45 x 10^{-18} Ci cm^{-2} s^{-1} (Wilkening et al. 1972). In a building with a concrete slab, radon from the soil can enter the indoor environment either by diffusion though the slab or by direct transport of soil gas though cracks in the slab. The radon flux from the slab due to diffusion though it is about an order of magnitude less than the flux from bare soil. Field measurements indicate significant radon transport though cracks and openings. These cracks can be microscopic and still be very effective radon transport conduits. The joints between the walls and the floor and loose fitting pipes though walls and floors adjacent to the soil substrate provide a direct pathway for radon to enter the building. Barometric pressure changes have a significant effect on radon exhalation; there is usually a doubling of radon exhalation (Kraner et al. 1964) when the barometric pressure falls 1%.

Radon from Building Materials

Masonry materials such as stone, concrete, and brick are often the primary sources of radon from building materials. Typical building materials contain < 2 pCi of ^{226}Ra.g^{-1} of material. However, there are situations where building materials that incorporate residues from industrial processes such as phosphate slag (Roessler et al. 1979) may contain ^{226}Ra in excess of 10 pCi.g^{-1} and may be a significant source of indoor radon. Building materials, such as wood and wood products, and nonsilicate material are not generally important because of their low ^{226}Ra content. In general, diffusion of radon from solid building materials is not a serious problem, except in well-insulated modern homes having an air exchange rate of < 0.1 per h.

Radon from Water

Radon released from water use indoors can be significant when the water comes from an underground acquifer in granite or other ^{226}Ra bearing rock. Radium-226 is usually relatively immobile in soils, but it can be dissolved in water and migrate from its source due to its relatively long half-life. In New England with ^{226}Ra content in granite of 2 to 5 pCi.g^{-1}, radon levels in water as high as 1,000,000 pCi.L^{-1} have been measured (Hess et al. 1979). In a home having radon in water of about 10,000 pCi.L^{-1}, the contribution to airborne radon will be about 1 pCi.L^{-1}. However, while radon in water is of great importance in a few regions, typical concentrations are well below 1,000 pCi.L^{-1}, and at this level water as a radon source can be ignored. Public surface water supplies usually contain very little dissolved radon and contribute < 0.06 pCi of radon in air.L^{-1} of water used (Bruno 1983).

FACTORS AFFECTING INDOOR RADON AND RADON DAUGHTERS

Like other indoor air pollutants, radon and radon daughter concentrations vary with location, time, and season. Generally, radon concentrations decrease from the basement to the upper floors or living quarters (George and Breslin 1980). Diurnal variations are found, with radon concentrations generally higher in the morning and lower during the afternoon. Barometric pressure, already mentioned, is also a cause of variation in radon concentrations. Seasonal variations in radon concentrations have been found but without any systematic pattern from one residence to another. They are different depending on the geographical area and on the type of heating used. During the heating season, when buildings are kept tight, there is a tendency for higher radon levels as opposed to lower levels measured during the warmer weather when there is an increase in ventilation or exchange with outside air.

Radon daughter air concentrations tend to follow the radon concentration, but they are reduced by deposition on surfaces caused by forced air systems and by filtration via the air conditioning or heating system. Family activity within the confines of a building is also responsible for variations in radon and radon daughter concentrations. Opening and closing doors and windows tends to decrease and increase radon daughter concentrations, respectively. House activities such as cooking, smoking, cleaning, and washing influence radon daughter concentration by increasing the concentration of airborne particles onto which radon daughters may become attached.

MEASUREMENT OF THE AIR CONCENTRATION OF RADON AND RADON DAUGHTERS

Because of the variation of radon and radon daughter concentrations within a building and within a locality, the exposure of the occupants of buildings should be assessed by measuring the air they breathe. This can be accomplished by grab, continuous, and integrated sampling.

Grab sampling is useful in some field applications such as for screening on a small scale, that is, for indicating whether a building requires detailed study or whether it can be considered acceptable (Lucas 1967; George 1976; Kusnetz 1956; Martz 1969; Thomas 1972). It is an attractive and powerful technique in guiding remedial action during mitigation work to reduce radon and radon daughters to acceptable levels. Grab sampling is not suitable for the assessment of the average indoor concentrations.

Continuous sampling is useful in scientific studies of the behavior of radon and radon daughters and for measuring real-time changes in their concentration, but it is unsuitable for large-scale surveys due to time and instrument complexity requirements and cost considerations.

The available data for indoor radon and radon daughter concentrations in the U. S. are inadequate to determine either an average or distribution of exposures. There is a need for a broader data base on human exposures. This can be accomplished by using long-term measuring techniques consisting of simple integrating monitors that measure the average exposure over several days, weeks, or months.

MEASUREMENT OF RADIATION EXPOSURE AND ESTIMATION OF DOSE

In planning a measurement program for radon and radon daughter airborne concentrations, the monitoring devices should be suitable for the particular application. There are several methods and monitors for measuring the average concentrations of radon and/or radon daughters. Since the inhalation exposure and consequent lung dose from the radon series (Table 1) arises from radon daughters, the appropriate choice is to use integrating devices that measure radon daughters or the so-called potential alpha energy emitted from airborne radon daughters. However, data from many typical residential buildings, where both radon and radon daughters were measured concurrently, indicate equilibrium factors between them ranging from 0.3 to 0.7 with an average value usually close to 0.5. Using this relationship, the radon daughter measurements, which are the most difficult to make, can be substituted with measurements of radon and appropriate corrections made.

A number of integrating radon monitors are available that measure the average concentration over days, weeks, or months, when repeated during different seasons. For screening purposes, there are simple passive radon detectors that are amenable to handling though the mail. Track etch detectors, suitable for monthly or longer periods of exposure, are commercially available (Alter and Fleischer 1981; Urban and Piesch 1981). The principle of radiation detection with track etch detectors consists of the damage imparted to the detector material by radon and radon daughter alpha particles. The advantages of track etch detectors are simplicity, low cost, and feasibility of use in extensive radon surveys. They are usually suited for quarterly measurements when repeated during different seasons of the year. Integrating times of 1 yr or more are possible if there is no urgent need for the results. Their main disadvantage is poor sensitivity for exposure periods of < 1 month.

For integrating time periods up to 1 week, the activated carbon detector is capable of measuring indoor radon with high precision and sufficient accuracy (Cohen 1983; George 1984; Gustafsson and Hildingsson 1984). The method uses simple detection principles for gamma rays emitted by the daughters of radon adsorbed on the carbon during exposure. The device is inexpensive and completely passive with no moving parts. It is ideal for screening surveys where data are needed in a short time (1 week or less). As with track etch detectors, the integrated measurements using activated carbon devices must be repeated during different seasons for the assessment of the average exposure. The main disadvantage is that the time period between the end of exposure and the analysis must be short (< 10 days). Field experience indicates that the detectors are suitable in surveys conducted on a regional basis where transit time is short.

Integrating detectors for measuring radon daughters in terms of working level are commercially available from several sources. They either use thermoluminescent materials (Schiager 1974; Guggenheim et al. 1979) or alpha particle counting techniques (Latner 1981). In both cases, the monitors are active, requiring power to move air though a filter by a pump and to operate the counting equipment. These monitors are too complex and costly to be used in large surveys. Their use is thus limited to scientific studies of the behavior of radon daughters and to serving as the basis for control strategies during remedial work.

The radiation dose to the respiratory system cannot be measured directly and is calculated by taking into consideration such factors as respiratory tract anatomy, radon daughter concentration, particle size, respiratory deposition, breathing characteristics, and occupancy. The factor for converting radon daughter exposure to absorbed dose to the bronchial epithelium developed by Harley and Pasternack (1982) is 0.7 rads.WLM^{-1}. Using a quality factor of 20 for alpha particle radiation, the absorbed dose is converted to dose equivalent. For example, the dose equivalent rate (rems.yr^{-1}) to the bronchial epithelium in 1 yr from an average radon concentration of 1 pCi.L^{-1} is calculated as follows:

1. Radon = 1 pCi.L^{-1}. Assuming the equilibrium ratio between radon daughters and radon is 0.5, then the PAEC = 0.005 WL.
2. (0.005 WL x 730 h.$month^{-1}$ x 12 months.yr^{-1})/(173 h.WM^{-1}) = 0.25 WLM.yr^{-1}.
3. The dose equivalent rate is 0.25 WLM.yr^{-1} x 0.7 Rads.WLM^{-1} x 20 = 3.5 Rems.yr^{-1}.

MEASUREMENT OF RADON SOURCES

Radon input into an existing building or the impact of radon on the indoor environment of a planned structure can be estimated at best by making measurements of radon exhalation from building materials, the underlying soil, and from the radium and radon content of the local water supply. The amount of radon entering the indoor environment depends on the fraction of diffusible radon, diffusion length and other transport mechanisms in the material. The radon exhalation rate can be measured by the rapid accumulation method or by the integrating technique. Rapid measurement of radon flux usually consists of an inverted can sealed against a surface for 1 to 5 h (Wilkening 1977). Samples of air under the accumulation can are transferred into radon detector devices for analysis (Lucas 1957; George 1976). Integrated measurements of radon flux up to 3 days are made with activated carbon detectors (Countess 1976; George 1984).

Measurement of the ^{226}Ra content of soil and building materials is made utilizing gamma-ray spectroscopy. Radon concentration in water, especially from underground sources, is measured by the liquid scintillation counting method (Pritchard 1977; Gesell 1977) and by the gamma-ray method (Countess 1978). Both techniques require water samples that are collected without degasing the radon.

SPECIAL MEASUREMENTS

Radon Daughter Particle Size and Respiratory Deposition

Since radon daughters contribute most of the radiation dose to the lung, it is important to know their size distribution, which ultimately determines the site of their deposition in the respiratory tract. Radon daughter particle size measurements are difficult to make, but they should be made in buildings representative of U. S. housing. In recent years, the Environmental Measurements Laboratory (EML) developed diffusion batteries suitable for measuring the size distribution of radon daughters in different environments (Sinclair et al. 1977; George et al. 1984; Knutson et al. 1984). Particle size measurements indicate a narrow size range of about 0.1 to 0.15 µm for the activity median diameter (George 1980).

Respiratory deposition of radon daughters is also difficult to measure using human subjects. Measurement results in the laboratory by George and Breslin (1967) and in residential buildings by Falk (1984) show an average total respiratory deposition of 30%.

Ventilation Measurements

Measurement of the air exchange in a building is very important because of its influence on the airborne concentrations of radon and radon daughters. The most commonly used method for measuring air exchange is the tracer gas technique (Sherman et al. 1980). This technique entails injection of a tracer gas into a building at one or more points and then monitoring the reduction of trace gas concentration with time. An alternate method is to inject the tracer gas at a low flow rate until a steady state concentration is achieved and then measure its concentration. Integrating techniques are under investigation exploring passive diffusion methods that yield a better estimate of the average air exchange. Ventilation measurements at present are not feasible on a large scale, but they can be very useful in diagnostic studies in selected buildings in conjunction with energy conservation upgrading or during mitigation work.

RADON AND DAUGHTER MEASUREMENTS IN U. S. BUILDINGS

The most important factor in assessing the exposure of occupants of buildings and in estimating the risk from natural radiation is the measurement of the airborne concentrations of radon and radon daughters. No systematic survey has been undertaken to estimate the exposure of the U. S. population. However, a substantial number of studies have been conducted in response to specific problems related to localities associated with uranium mill tailings, uranium processing wastes, phosphate mining, uranium mineralized lands, and most recently to energy conservation programs in ordinary buildings. Because of the manner in which data were collected in the U. S., the National Council on Radiation Protection and Measurements (NCRP) could not estimate the number of U. S. buildings exceeding the recommended remedial action level for radon daughter exposures (Action Level = 0.04 WL or 2 WLM.yr^{-1}).

Typical measurements made either directly by EML or in cooperation with other agencies over a period of 10 yr are listed in Table 2. The data were obtained in different geographical areas, during different seasons, in the living room, dining room, bedroom, or family room of 466 residential buildings. For dosimetric purposes, the integrated and grab sample measurements of radon have been converted to WL in accordance with Equation 1. The distributions of radon daughter concentrations are approximately lognormal. The results show that the average indoor airborne concentrations of radon and radon daughters vary substantially in different geographical areas. This may not be surprising because the surveys were conducted in various types of buildings, constructed over various underlying geological substrates, with and without energy conservation upgrading.

The distribution of airborne radon concentrations obtained in 1377 homes in 21 states including those listed in Table 2, compiled by Nero, have been found to be log normal with a geometric mean concentration of 0.9 pCi.L^{-1} and a geometric standard deviation of 2.8 with an average value of 1.5 pCi.L^{-1}. If the measurements in these homes are representative of U. S. housing, it appears that radon and radon daughter levels in $\sim$ 1 million homes exceed the recommended action level value of 8 pCi.L^{-1} or 0.04 WL. The data in Table 2 suggest that a substantial fraction of the homes surveyed fall into this category.

CONCLUSIONS

Indoor airborne levels of radon and radon daughters in some buildings may account for some of the lung cancers in the U. S. The adverse health effects from indoor radon and radon daughters are compounded by the implementation of energy conservation measures and the introduction of new building technology and materials. Radon levels vary from one area to another, indicating geographical inhomogeneity in radon source strength.

The estimated risk from what is considered average background indoor radon daughter exposure (0.2 WLM.yr^{-1}) is about 1/20th of the present occupational limit, and the observed distribution is such that a substantial fraction of the general population is exposed to levels as high as one half of the occupational limit. The lifetime lung cancer risk per million individuals exposed to 0.2 WLM.yr^{-1} (background level) and 2 WLM.yr^{-1} (the action level) is 1800 and 18,000 with annual mortality rates of 10,000 and 100,000, respectively, in the U. S. population (NCRP 1984).

The measurements for radon and radon daughters in the U. S. suggest that there are anomalous regions where the soil radon entry rate is well above average. The most effective and lasting control measures appear to be finding the source of radon and preventing its entry indoors. If that is not practical, then radon and daughter concentrations can be reduced by increasing the ventilation rate. With respect to new housing, radon levels can be controlled by using the proper design. In the pursuit of energy conservation, it is apparent that more research is needed on radon sources and controls in order to reduce the health risk associated with increased exposure to indoor radon and radon daughters. Since there are no federal guidelines and standards for indoor radon exposure, it is prudent to follow the NCRP recommendation: that continuous exposure of individuals in the general population to 2 WLM.yr^{-1}, or greater, of radon daughters should be avoided.

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TABLE 1

Characteristics of Radon Decay Series

Isotope	Alpha Ray Energy (MeV)	Half-Life	No. of Atoms.pCi^{-1}	Ultimate Alpha Ray Energy. Atom^{-1}	Total Alpha Ray Energy (MeV.pCi^{-1})
^{222}Rn	5.49	3.82 days	17,700	Insignificant	None
^{218}Po	6.00	3.05 min	10	6.00 + 7.68	137
^{214}Pb	0	26.8 min	86	7.68	660
^{214}Bi	0	19.7 min	63	7.68	484
^{214}Po	7.68	10^{-6} min	8 x 10^{-6}	7.68	0.00006
^{206}Pb (Stable)				TOTAL =	1,281[a]

[a] The value of 1281 MeV.pCi^{-1} = 0.01 WL.

TABLE 2

Radon Daughter Concentrations in Residential Living Areas in 13 U. S. Localities

Location of Buildings	No. of Buildings	Geometric Mean WL	WLM.yr^{-1}	Dose Rate Equivalent to Epithelial Cells (REM.yr^{-1})
NY City Area	11	.0023	.12	1.7
Lewiston, NY	10	.0026	.13	1.8
New Jersey	9	.0068	.34	4.8
Middlesex, NJ	15	.0026	.13	1.8
Chester, NJ	21	.0080	.40	5.6[a]
Philagelphia Area	31	.0092	.46	6.5[b]
Canonsburg, PA	8	.0047	.24	3.4
Eastern, PA	36	.0165	.84	11.8[c]
Damascus, MD	41	.0152	.77	10.8[d]
Central Florida	29	.0028	.14	2.0
Grand Junction, CO	62	.0070	.35	4.9
Oak Ridge, TE	14	.0064	.32	4.5
Butte, MT	179	.0173	.88	12.3
Occupational Limit			4.00	56.00

[a] 5% of homes have radon concentrations = or > than action level.
[b] 16% '' '' '' ''
[c] 30% '' '' '' ''
[d] 10% '' '' '' ''

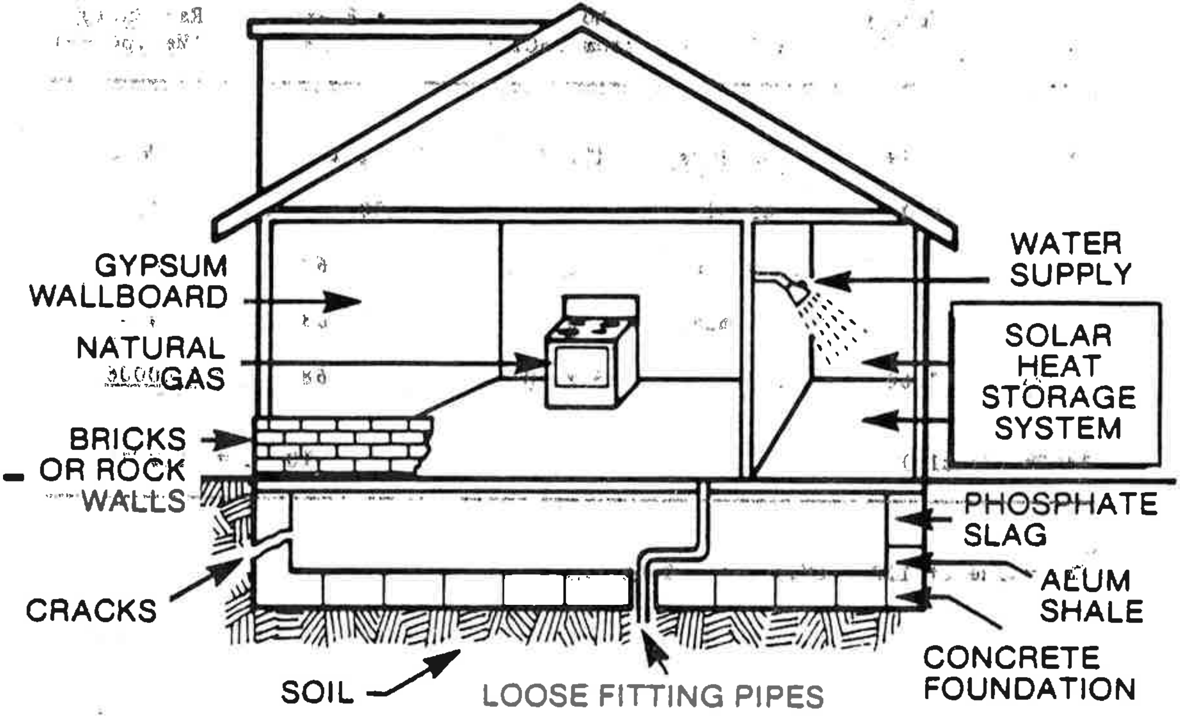

Figure 1. Sources and pathways of radon in a residential building

[illegible]