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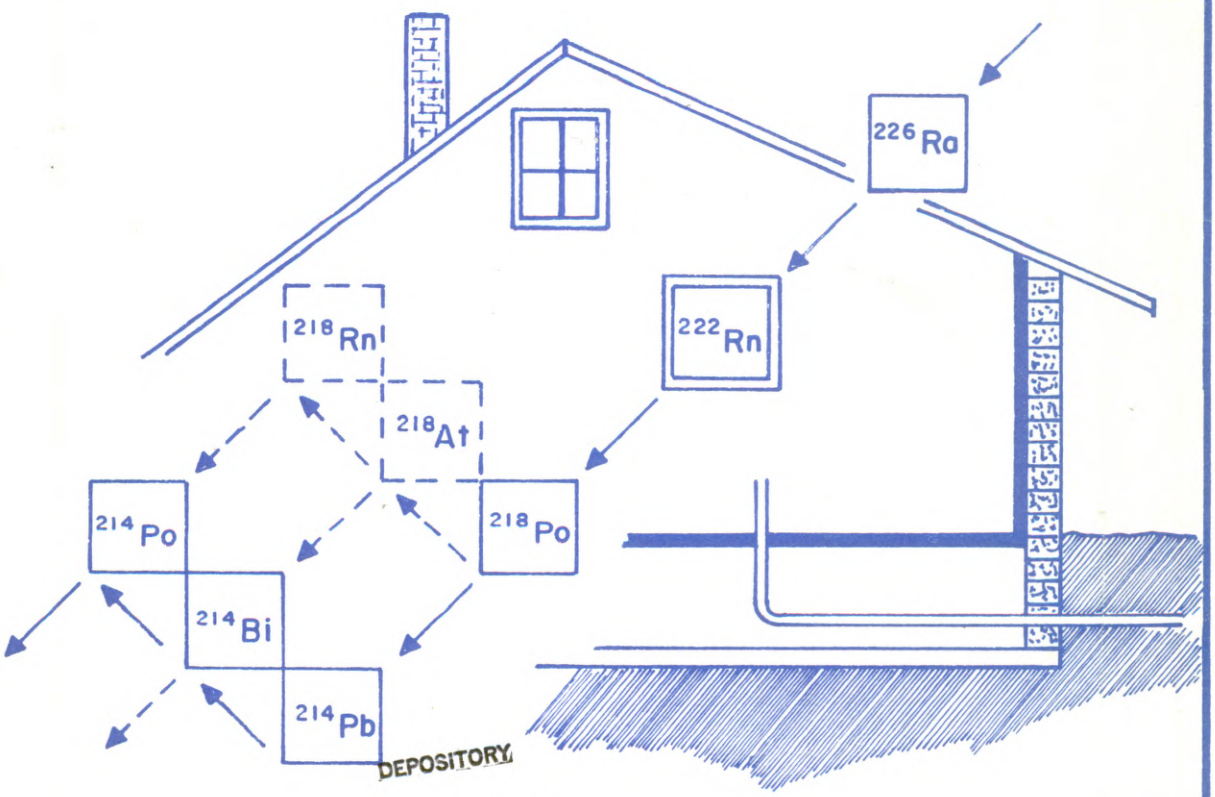
Radon in Buildings



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Radon in Buildings

Proceedings of a Roundtable Discussion of Radon in Buildings held at the National Bureau of Standards, Gaithersburg, Maryland, June 15, 1979

Edited by

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PREFACE

During the past few years, potential health hazards of radiation exposure due to radon in buildings have received increasing attention. Many studies have indicated that indoor concentrations of radon and its radioactive-daughter products are frequently higher than outdoor concentrations. At the same time, there is an increasing interest in energy conservation in buildings including strategies such as reducing air exchange rates which may result in elevated radiation levels. It is becoming clear that radiation protection and building technology are in some cases interdependent. Radiation protection should be considered in building designs; similarly, known building technologies should be considered in the control of elevated indoor radon levels.

As a result of these concerns, a Roundtable Discussion of Radon in Buildings, jointly organized by the NBS Center for Radiation Research and the Center for Building Technology, was held June 15, 1979 at the National Bureau of Standards in Gaithersburg, Maryland. Participants included individuals with programmatic as well as scientific and technical expertise in radiation protection, radiation measurement and building technology areas. They represented a wide range of private and government institutions with diverse radiation and building technology interests.

The objective of the meeting was to bring together these participants to exchange information and to draw attention to some of the problems and research needs, and provide a forum for a general discussion of radon in buildings. Topics that were considered include: (1) the sources and pathways of radon in buildings; (2) diffusion and interaction properties and the distribution of daughter products on aerosols; (3) the biological and health effects; (4) measurement methods; and (5) strategies to minimize indoor levels of radon, such as increased ventilation and selection and treatment of building materials.

The program consisted of five review papers, twenty brief reports from some of the participants on their current research activities and interests, and the roundtable discussion. This publication contains the review papers, summaries of the brief reports, and edited summary of the roundtable discussion. The papers and summaries are printed in the order in which they were presented. The roundtable discussion session of the meeting was recorded for subsequent analysis and use in preparing the summary. Apart from a few minor changes, the papers and summaries are essentially those submitted by the speakers as the final versions of their presentations. The editors apologize for any inadvertent mistakes. For additional information on the presentations, contact the speakers directly.

Certain commercial equipment, instruments and materials are identified in this report in order to adequately specify experimental procedure. In no case does such identification imply recommendation or endorsement by the National Bureau of Standards, nor does it imply that the material or equipment identified is necessarily the best available for the purpose.

The roundtable discussion and this subsequent report would not have been possible without the guidance and encouragement of Elmer H. Eisenhower. The editors also gratefully acknowledge the excellent secretarial assistance of Karen Fritz, Jennifer Wright and Edythe Kramer in organizing and arranging the meeting and in preparing this report, and the assistance of the National Bureau of Standards Technical Information and Publications Division in the preparation and publication of this report. Lastly, the editors wish to thank the authors and participants of the roundtable discussion for their enthusiasm, lively discussion, and contributions to this report.

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ABSTRACT

This is the proceedings of a Roundtable Discussion of Radon in Buildings held June 15, 1979 at the National Bureau of Standards in Gaithersburg, Maryland. The meeting brought together a number of participants with diverse interdisciplinary interests in radiation protection, radiation measurement and building technology, provided a forum to exchange information, and drew attention to some of the problems and research needs associated with radiation exposure due to radon in buildings. Emphasis was placed on (1) the characterization of the sources and pathways of radon in buildings; (2) the biological and health effects; (3) measurement considerations; and (4) strategies and control technologies to minimize indoor radiation exposure.

Key Words: Buildings; environment; health; measurements; radiation; radon; radon daughters; ventilation.

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THE PHYSICS AND INTERACTION PROPERTIES OF RADON AND ITS PROGENY

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This review summarizes the elementary physical and chemical properties of radon and its radioactive decay products. These properties govern their behavior in the environment and in buildings where they may concentrate and create potential human health hazards. Radon and its progeny, as intermediate decay products in the naturally-occurring long-lived primordial radioactive series, are characterized by a unique and complex behavior. The characteristics and properties are considered in describing the natural and "technologically enhanced" sources of radon, the radioactive decay relationships involved, the type and energy of the radiations emitted in the decay of radon, and the interactions of radon and its progeny in natural and building environments.

The element radon is a noble gas which has three naturally-occurring isotopes with mass numbers of 219, 220 and 222. All three are radioactive and have short halflives: ^{219}Rn, 3.96 seconds; ^{220}Rn, 55.6 seconds; ^{222}Rn, 3.82 days. Historically, they were named "actinon", "thoron" and "radium emanation", respectively[1]. The three radon isotopes occur in nature as intermediate decay products in the three radioactive series headed by the long-lived primordial radionuclides uranium-235 ("actinium"), thorium-232 and uranium-238. The ^{232}Th and ^{238}U decay series are illustrated in Figures 1 and 2. Natural uranium consists of 99.27 percent (by weight) ^{238}U and 0.73 percent ^{235}U. For this reason, the ^{235}U series is less important. Additionally, the decay products of ^{235}U, i.e., the subsequent radionuclides of the series (sometimes called

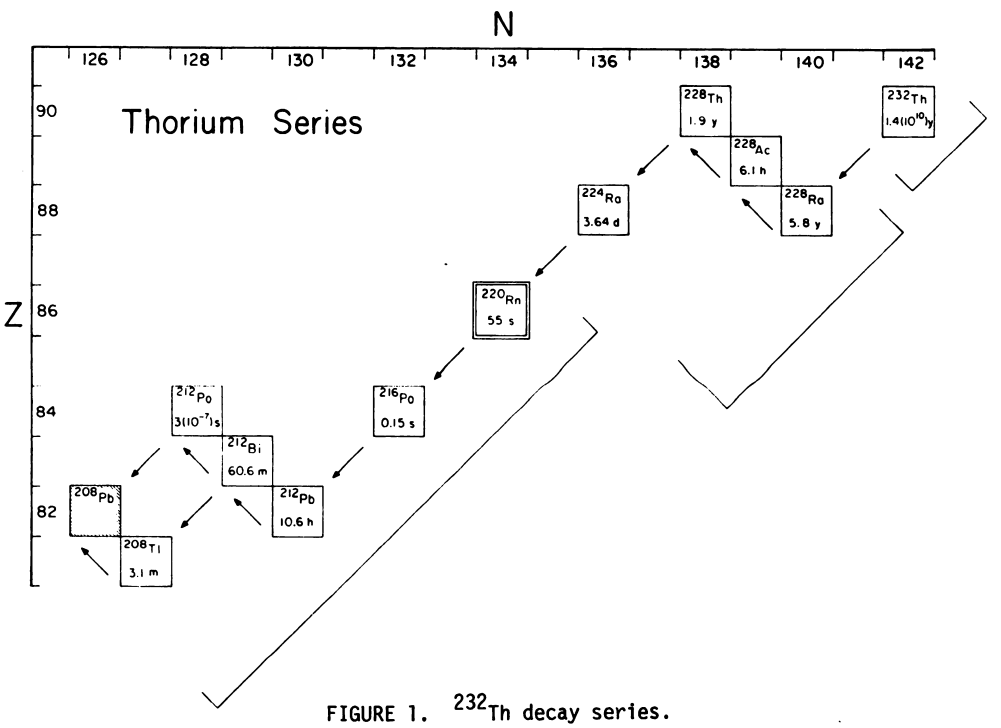

FIGURE 1. ^{232}Th decay series.

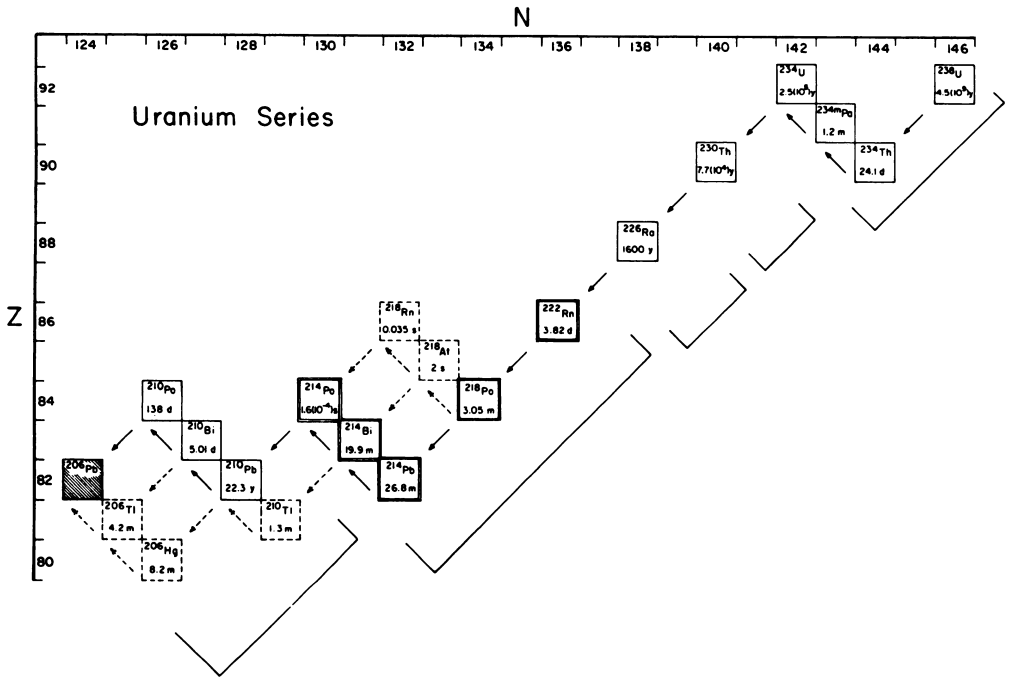

FIGURE 2. ^{238}U decay series.

decay "daughters" or "progeny"), have relatively short halflives and do not appear in the environment in significant concentrations[2]. Natural thorium consists entirely (100% abundance) of the isotope ^{232}Th. Therefore, both the decay series of ^{232}Th and ^{238}U are of importance as natural sources of radon.

The complexity of the naturally-occurring radioactive series arises because every isotope of every element with an atomic number greater than 83 (bismuth) is radioactive and decays primarily by either alpha or beta decay. Alpha decay (α) decreases the atomic number (Z) by two and the mass number (Z + N) by four units. Beta decay (β^-) increases the atomic number by one, but does not change the mass number (see Figures 1 and 2). Thus, the nucleus ^{232}Th with an atomic number of 90 and mass 232 emits 6 alpha particles and 4 β^- particles before it achieves stability and results in the stable lead isotope ^{208}Pb. This progression toward stability illustrated in Figures 1 and 2 is further complicated by branching decay so that the complete series, ignoring isomers, consist of 12 distinct nuclear species for the ^{232}Th series, and 20 for the ^{238}U series.

In alpha decay, the nucleus emits one or more α particles with discrete energies. Usually, an α particle of one energy value predominates. In β^- decay, on the other hand, the nucleus emits β^- particles which have a continuous spectrum of energies, ranging from near zero to a maximum value which is characteristic of the particular radionuclide. In either case, the decaying nucleus may not release all of the available decay energy, but may leave the product nucleus in an excited state from which it decays by some form of electromagnetic transition which is primarily by the emission of gamma rays. Thus, the decay of radionuclides in a naturally-occurring series may involve the emission of many different α particles, β^- particles and gamma rays. For example, the decay of the ^{222}Rn subseries shown in Figure 3 comprises three major and at least four other less intense α-particle transitions, over 50 different β^--particle transitions, and nearly 200 distinct gamma-ray transitions. The major radiations, their energies and intensities

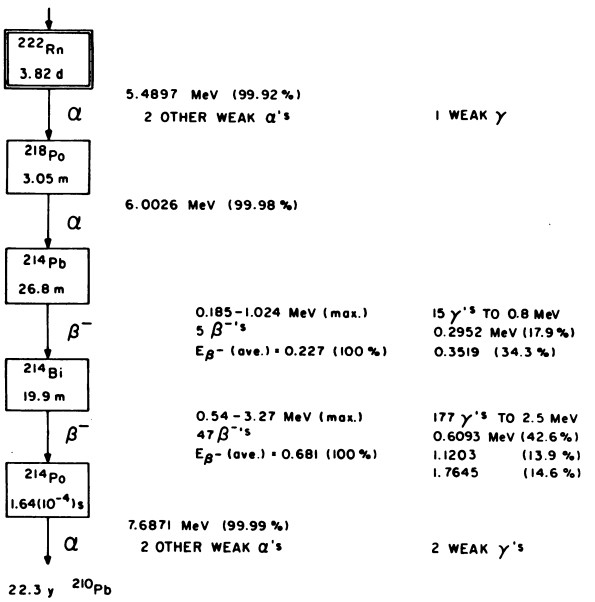

FIGURE 3. ^{222}Rn decay subseries.

for the naturally-occurring radioactive series have been summarized in a number of NCRP Reports [3, 4]. More complete tabulations are also available [5, 6].

If undisturbed by either chemical or physical processes, the radionuclides in a decay series attain a state of radioactive "equilibrium". Unlike chemical equilibrium, this state does not imply reversibility of reactions, nor does it describe a true "steady state" condition since the concentrations of the radionuclides continually change. In the case of a long-lived parent and short-lived daughter, equilibrium is achieved when the ratio of the decay rates of parent and daughter becomes constant. Contrary to what might be expected intuitively, how fast equilibrium occurs depends primarily upon the halflife of the daughter activity, and only secondarily on the halflife of the parent. Once equilibrium is established, however, both activities decay with the parent's halflife. In equilibrium, the daughter activity will be greater than the parent activity by the factor $T_1/(T_1-T_2)$ where T_1 and T_2 are the parent and daughter halflives, respectively. In the naturally-occurring radioactive series, at equilibrium, the decay rate of every radionuclide in a given series is very nearly equal to the decay rate of the radionuclide that heads the series.

The radionuclides in the ^{238}U and ^{232}Th series, however, rarely attain a state of equilibrium in the environment. The shorter-lived daughter radionuclides are more directly controlled by chemical or physical processes than by the halflife controlled genetic decay relationships. A state of "disequilibrium" in a decay chain occurs when the activities of the various radionuclides in the series exist in some arbitrary nonequilibrium ratio to each other. Such situations can arise when parent and daughter activities are chemically and physically different, and can be separated in nature by ordinary geological processes. This can include the selective leaching of minerals by ground water, the diffusion of radon, partitioning of substances in mixtures, as well as differences in electrostatic attraction, physical adsorption, and containment within minerals, rocks and soils.

The uranium series, headed by ^{238}U (Figure 2) is generally separated into five subseries [7] as a result of the different chemical and physical properties, and the sufficiently long halflives of the intermediate members. The members of the various subseries are designated in Figure 2. Within each subseries, the activities are generally near a state of radioactive equilibrium with the head of the subseries. Brief descriptions of the ^{238}U subseries, adapted from NCRP No. 45 [7] follow:

(i) The first subseries consists of the two uranium isotopes ^{238}U and ^{234}U and the two intermediate radionuclides, ^{234}Th and ^{234m}Pa. The halflives of the intermediate members are too short to allow significant separation from the uranium isotopes. Although isotopic fractionation of uranium can occur in some soils and in sea water, ^{238}U and ^{234}U may be considered to be in radioactive equilibrium.

(ii) ^{230}Th comprises the second subseries. The relative chemical inertness of tetravalent thorium compounds compared to the tendency of uranium to exist in hexavalent oxidation states, and long ^{230}Th halflife permits separation from the uranium precursors. Significant disequilibrium can occur in oceans where thorium precipitates and is deposited in sedimentary layers.

(iii) The third subseries is composed of ^{226}Ra. It is frequently distributed differently than its precursors since radium is relatively mobile with respect to uranium and thorium. Except for certain ^{226}Ra "hot spots" of radium sulfate remaining from uranium deposits, radium rarely concentrates and is widely dispersed.

(iv) The fourth subseries includes ^{222}Rn and its progeny ^{218}Po, ^{214}Pb, ^{214}Bi, and ^{214}Po. Although radon accompanies radium in nature, as a noble gas, it can diffuse and migrate considerable distance from its parent. Radon is spontaneously liberated in varying extents from crystalline compounds and other solids containing radium. Relatively impervious minerals such as zircon lose very little of the radon formed in them, while open-lattice structures may lose most of their radon under natural conditions. The degree (fractional amount of radon that is missing) to which a mineral loses radon is called the "emanating power." Similarly, the rate of loss of radon from materials is called the "emanation rate". Radon is soluble in water and can be found in concentrations much greater than its radium parent. The short-lived decay products with halflives ranging from 26.8 minutes to 164 microseconds are readily ionized and attach to surfaces such as dust partices in air. Undisturbed, the subseries reaches radioactive equilibrium within a few hours.

(v) The fifth and final subseries starts with the long-lived ^{210}Pb and continues through ^{210}Bi and ^{210}Po where it finally terminates in stable ^{206}Pb.

The ^{232}Th series (Figure 1) has also been described in NCRP Report No. 45 [7]. The series is characterized by the long-lived ^{232}Th parent, and progeny which are all relatively short-lived. It may be considered in three subseries consisting of

(i) ^{232}Th itself, the least mobile radionuclide in the series;

(ii) the two radium isotopes ^{228}Ra and ^{224}Ra and the two intermediate members ^{228}Ac and ^{228}Th; and

(iii) the seven-radionuclide sequence starting with the inert noble gas ^{220}Rn and terminating in stable ^{208}Pb.

Generally, disequilibrium within the series does not occur to a large extent. Radioactive equilibrium occurs in the geologically short time of about 60 years if the members, particularly radium, do not migrate. Disequilibrium situations arise mainly at interfaces between soils, natural waters, natural gas and petroleum deposits, and the atmosphere.

The short halflife of ^{220}Rn (55 seconds) precludes significant migration from its natural sources. Therefore except in special situations where thorium-rich minerals like zircon and monazite are concentrated in placer deposits[8] such as sands on the Florida coast, ^{220}Rn (from the thorium series) is not as important as ^{222}Rn (from the uranium series) as a natural source of radon exposure. For this reason, the remainder of this review will focus on the ^{222}Rn subseries.

The naturally-occurring radionuclides in the ^{222}Rn subseries, whether undisturbed in the ambient background or technologically enhanced, can result in both external and internal exposure to human populations. As indicated in Figure 3, decay of the ^{222}Rn subseries radionuclides involves emission of both α and β^- particles as well as gamma radiation. For external exposure, the radionuclides of primary importance are ^{214}Pb and ^{214}Bi. In fact, the gamma rays of ^{214}Bi are the most abundant and energetic of the

uranium series. The greatest contribution to the external exposure is made by gamma radiation from these sources. The external radiation from α and β^- decay does not contribute significantly to the absorbed dose in the tissues and organs of man. Internal exposure, which is generally more hazardous, may arise, however, from inhalation or ingestion of the radionuclides. The major contribution to the internal dose comes from airborne α-emitting members of the ^{222}Rn subseries which account for about 45% of the alpha energy of the uranium series. Ambient air can be enriched in ^{222}Rn and its progeny because of emanation of the gaseous radon and its subsequent decay in the atmosphere. These radionuclides can be inhaled and contribute to the absorbed dose in lung tissue. Although the dose to the lung is most significant, the ^{222}Rn progeny are isotopes of lead, bismuth and polonium and can be translocated from the lung to other organs of the body [9].

Since the exposure to man from ^{222}Rn and its progeny will depend on the concentration and distribution of these radionuclides in man's environment, one must consider the radioactive decay and growth relationships involved in the ^{222}Rn subseries. The general mathematical forms of the decay and growth equations governing radioactive-series decay are often referred to as the "Bateman equations" [10]. The derivations are available in many standard texts [11], and only the results are summarized here.

The decay of a single radionuclide is given by

$$A(t) = A^{\circ} e^{-\lambda t}$$

where A(t) is the activity of the radionuclide at any time t, A° is its activity at time t=0, and λ is the decay constant given by $\lambda = \ln 2/T$ where T is the characteristic halflife of the radionuclide. The activity of a radioactive daughter produced from decay of its parent is

$$A_2(t) = A_1^{\circ}\left[\frac{\lambda_2}{\lambda_2-\lambda_1}\right]\left(e^{-\lambda_1 t} - e^{-\lambda_2 t}\right) + A_2^{\circ} e^{-\lambda_2 t}$$

where subscripts 1 and 2 refer to parent and daughter, respectively. The second term in the above equation is that part of the daughter activity originally present at time t=0 which is still present at time t. If no daughter is present initially ($A_2^{\circ}=0$), this term is zero. The activity of a radioactive granddaughter can be expressed similarly. If parent 1 decays through daughter 2 to produce granddaughter 3, then (for the case of $A_2^{\circ}=A_3^{\circ}=0$) the granddaughter activity is

$$A_3(t) = A_1^{\circ}\left\{\frac{\lambda_2\lambda_3 e^{-\lambda_1 t}}{(\lambda_2-\lambda_1)(\lambda_3-\lambda_1)} + \frac{\lambda_2\lambda_3 e^{-\lambda_2 t}}{(\lambda_1-\lambda_2)(\lambda_3-\lambda_2)} + \frac{\lambda_2\lambda_3 e^{-\lambda_3 t}}{(\lambda_1-\lambda_3)(\lambda_2-\lambda_3)}\right\}$$

The above equations may be expressed generically for the $n^{\underline{th}}$ generation activity as

$$A_n(t) = A_1^{\circ}\sum_{i=1}^{n} C_i\, e^{-\lambda_i t}$$

where

$$C_i = \frac{\prod_{k=2}^{n}\lambda_k}{\prod_{j=1}^{n}(\lambda_j-\lambda_i)} \qquad (j\neq i).$$

For an initially pure source of ^{222}Rn, Figure 4 shows the growth and decay curves of the various members of the ^{222}Rn subseries computed from these equations. The daughter ^{218}Po grows in and reaches equilibrium with ^{222}Rn within about one half hour. The granddaughter ^{214}Pb

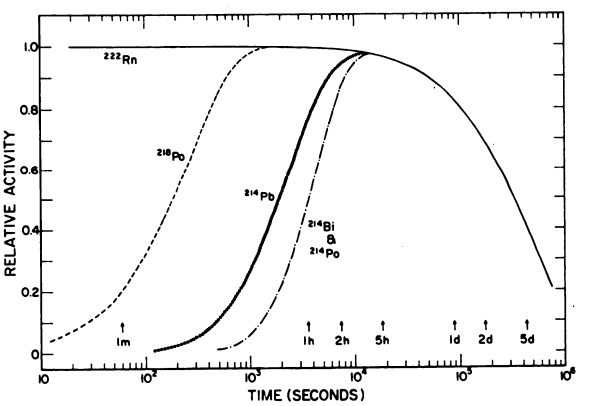

FIGURE 4. Decay of an initially pure source of Rn-222, and the subsequent growth and decay of its progeny.

achieves equilibrium within several hours, and is followed shortly thereafter by the ^{214}Bi and ^{214}Po progeny. As indicated earlier, the approach to equilibrium is controlled mainly by the half-lives of the progeny. Once equilibrium is achieved, the ^{222}Rn and progeny decay with the halflife of ^{222}Rn. From Figure 4 it is easy to see that disequilibrium activity ratios of progeny to ^{222}Rn will occur if a disturbing influence removes or depletes some of the ^{222}Rn progeny prior to the establishment of equilibrium.

Similarly, the growth and decay of the total α- and β^-- particle emission rates for an initially pure source of ^{222}Rn is illustrated in Figure 5. The decay of the ^{222}Rn subseries results in the ultimate emission of three α and two β^- particles for each ^{222}Rn radioactive decay. As a result, an undisturbed ^{222}Rn source of unit activity (s^{-1}) will achieve a maximum total α-emission rate approaching nearly 3 $\alpha \cdot s^{-1}$, and a maximum total β^--emission rate of nearly 2 $\beta^- \cdot s^{-1}$. Once equilibrium is achieved, the α- and β^--emission rates decrease with the half-life of ^{222}Rn. As before, the emission rates will also be altered by any perturbation or disturbing influence that removes or depletes the ^{222}Rn progeny.

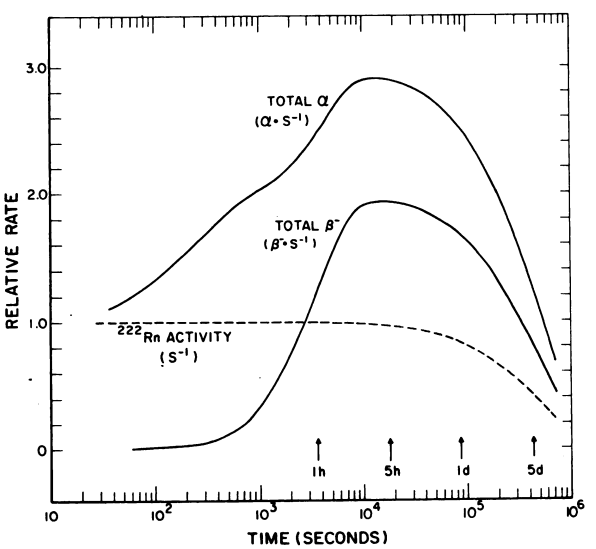

FIGURE 5. Growth and decay of the total α-and β^--particle emission rates for an initially pure source of Rn-222.

The dose to the human lung resulting from exposure to radon is predominately attributed to the α particles emitted in the decay of the ^{222}Rn progeny, and not that from the radon itself. At the same time, the occurrence of nonequilibrium mixtures of ^{222}Rn and progeny is common.[12-14] As a result, a unit of measurement called the "working level" (WL) was introduced in the late 1950s which takes account of the importance of the ^{222}Rn - progeny concentrations and the likely absence of radioactive equilibrium between ^{222}Rn and its progeny. The WL unit, which is now widely used for radiation protection, control and regulatory purposes, is defined as

> any combination of the short-lived radon progeny (^{218}Po, ^{214}Pb, ^{214}Bi and ^{214}Po) in one liter of air that will result in the ultimate emission of 1.3×10^5 MeV of alpha - particle energy.[15-16]

The numerical value of the WL (1.3×10^5 MeV per liter) is derived from the total α-particle energy ultimately emitted in the decay of the short-lived ^{222}Rn progeny that are in radioactive equilibrium with 100 picocuries of ^{222}Rn per liter of air. Table 1, adapted from Evans[16], illustrates the definition of the WL unit and the relevant numerical data. Although the numerical value of the WL unit is derived with the assumption of radioactive equilibrium (i.e., 100 $pCi \cdot L^{-1}$ of each ^{222}Rn progeny), the unit is applicable for any mixture of short-lived ^{222}Rn progeny including nonequilibrium situations. It is also important to remember that the WL is a concentration unit based on one liter of air which may be subject to density variations. The definition, interpretation and shortcomings of the WL unit have been excellently described by Evans[16]. When considering radiation exposure to the human lung from radon sources, the WL unit is presently the most useful single-parameter measure of the effective airborne radioactivity.

The relative contributions of the various progeny to the WL in an initially pure source of ^{222}Rn is shown in Figure 6. This growth in the WL can be compared to the growth of the ^{222}Rn progeny (Figure 4). The earliest contribution

TABLE 1

Definition of the "Working Level" (WL) Unit.

Nuclide	α-Particle Energy (MeV)	Half-Life	Number of Atoms per 100 pCi	Ultimate α-Particle Energy Per Atom (MeV)	Total Ultimate α-Particle Energy Per 100 pCi (MeV)	Percent of Total α-Particle Energy
^{222}Rn	5.490	3.8235 d	$1.763(10^{6})$	excluded	--	--
^{218}Po	6.003	3.05 m	$9.768(10^{2})$	6.003 +7.687	$0.1337(10^{5})$	10.4%
^{214}Pb	0	26.8 m	$8.583(10^{3})$	7.687	$0.6598(10^{5})$	51.4%
^{214}Bi	0	19.9 m	$6.374(10^{3})$	7.687	$0.4900(10^{5})$	38.2%
^{214}Po	7.687	$1.643(10^{-4})$s	$8.770(10^{-4})$	7.687	$0.0000(10^{5})$	0.0%
				TOTAL	$1.2835(10^{5})$	100.0%
				CALLED	1.3×10^{5} MeV	

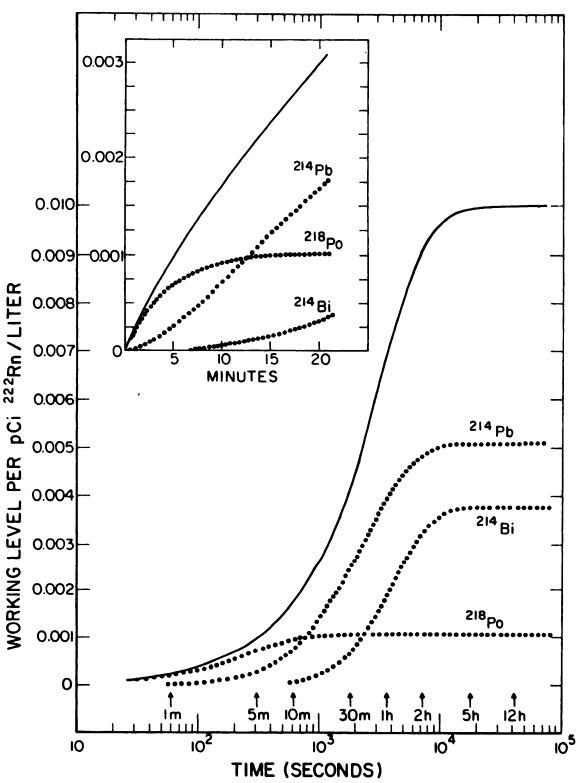

FIGURE 6. Growth of the WL for an initially pure source of Rn-222, and the relative contributions to it from the progeny.

to the WL is made by ^{218}Po, followed by ^{214}Pb, and finally by ^{214}Bi. A significant contribution to the WL by ^{214}Po is precluded by its extremely short halflife (Table 1). If the initially pure ^{222}Rn source remains undisturbed, the largest contributions to the WL are made by ^{214}Pb and ^{214}Bi which contribute 51% and 38% of the ultimate α- particle energy, respectively. As indicated in Figure 6, these relative contributions by the various progeny (last column, Table 1) will only be achieved after several hours. They may never be achieved if the radon is dispersed (such as by rapid air changes) before sufficient time has elapsed for the build-up of the progeny, or if the progeny are removed from the air (such as by filtration). In these situations, more significant contributions to the WL may be made by the first decay product, ^{218}Po. For example, Evans [16] compared the relative contribution of the ^{222}Rn progeny to the total α-particle energy for air of various "ages". The results for "3 min air" (i.e., a ^{222}Rn air sample that has had only 3 minutes in which to accumulate its radon progeny) and "10 min air" is compared to "equilibrium air" in Table 2.

TABLE 2

Percent of total α-particle energy contributed by the progeny of ^{222}Rn for various "air ages".

	^{218}Po	^{214}Pb	^{214}Bi
"3 min air"	83%	17%	0.5%
"10 min air"	54%	42%	4%
"equilib. air"*	10%	51%	38%

*See Table 1.

Before considering the factors influencing and causing these variations, as well as the important characteristics and interaction properties of the ^{222}Rn progeny, it may be useful to summarize the major sources of radon in the environment and in buildings.

Since very little radium is found in the atmosphere, essentially all radon is considered to originate from natural or disturbed terrestrial sources.[9,17] Radon, which originates from the decay of radium in soils, rocks and minerals, diffuses and emanates more or less rapidly depending on the porosity, and can become widely distributed before it decays. The vertical distribution and occurrence of ^{222}Rn and its progeny at various altitudes has been measured by many investigators (Cf., 1977 UNSCEAR report[17]). In general, radon concentrations decrease with increasing height. Based on a review of nearly 1000 measurements of the ^{222}Rn emanation rate from soils of various types at locations across the earth, Wilkening, et al[18] obtained a world-wide overall mean of 0.42 $pCi \cdot m^{-2} \cdot s^{-1}$ with a range from 0.006 to 1.4 $pCi \cdot m^{-2} \cdot s^{-1}$.

The emanation rate is influenced by the type, porosity, moisture content and temperature of the soil, and the atmospheric pressure and thermal stability of the ground air[19-25]. The rate is reduced when the ground is cold or frozen, when wet following heavy rainfall, by snow cover, and by increasing barometric pressure. Diurnal variations in the emanation rate are frequently observed with maxima at night and minima in the afternoon. The variations, which can range up to 100%, are caused by two opposing factors.[18] At night, emanation rate is increased because of upward convective flow which results from the temperature variations in the top soil layer. It can also be increased during daylight hours because of the greater turbulent mixing in the ground atmosphere. As a result, the diurnal fluctuations are considerably smoothed. Seasonal variations of the emanation rate are controlled by climatic conditions (snow cover, temperature, thermal stability, etc.).

The total world-wide release of ^{222}Rn has been estimated as 50 $Ci \cdot s^{-1}$ by Wilkening[18] and 75 $Ci \cdot s^{-1}$ by Harley[26], which leads to a total equilibrium activity of 25 to 40 million curies. Harley[26] has also calculated that this corresponds to an average surface air concentration of 70 $pCi \cdot m^{-3}$, and about 100 $pCi \cdot m^{-3}$ in the northern hemisphere. These values are in reasonable agreement with surface air measurements of ^{222}Rn concentrations (and those inferred from measurements of ^{222}Rn progeny concentrations). Average outdoor concentrations of ^{222}Rn are typically between 100 and 200 $pCi \cdot m^{-3}$[9,17,18]. in the northern hemisphere.

In addition to the truly natural sources, exposure to radon and its progeny may be enhanced by human activities or technology that redistributes the naturally-occurring terrestrial radionuclides. This source category, which has been termed "technologically enhanced natural radioactivity" (TENR)[27], is used to distinguish controllable natural radioactivity sources from those generally uncontrollable. Examples of activities that enhance naturally-occurring radiation include mining operations, well drilling and development, burning of fossil fuels, and the use of construction material containing radionuclides (e.g., granite, brick, concrete block). A number of these TENR sources of radon have been extensively studied over the past few years, and some will be briefly highlighted.

Coal[33-35]

Coal contains varying concentrations of

uranium, thorium and their radioactive progeny. In the U.S., the coal which is mined in the western states contains significantly higher amounts of uranium (0.001 to 0.1%) than coal mined in the north central and eastern regions (<0.001%).[28-30] Exposure to radon and its progeny can occur in several ways, one of which is during the mining process. Radon is released into the atmosphere during the exposure of coal seams as well as during the breakup of the coal being mined. In this case, the concentration of radon in the mine atmosphere depends not only on the uranium and thorium content of the coal, but also on the ventilation rate of the mine. The dominant area of radon emission from coal, however, occurs at power generating plants during the combustion process. With the exception of radon, which is released as a gas into the atmosphere, all other radionuclide emissions from coal combustion are associated with the discharge of fly ash.[32] This fly ash may contain radon progenitors, such as uranium or radium, which ultimately decay to radon. Other possible sources of radon dispersal include run off from fly ash storage and disposal areas, and the use of fly ash in construction material.[28]

Natural Gas[33-35]

Radon which is found in the same geological strata where natural gas production sites are located, diffuses from the strata into the wells where it is collected along with the natural gas. Based on a number of selected sites throughout the U.S., Johnson, et. al.[34] reported an overall average ^{222}Rn concentration in natural gas at production wells of 37 $pCi \cdot L^{-1}$. The natural gas processing and distribution network plays an important role in reducing the concentration of radon. The concentration is greatest at the wellhead and decreases with distance. In selected distribution lines, Johnson, et. al.[34] reported an overall average ^{222}Rn concentration of 23 $pCi \cdot L^{-1}$. Natural gas used in unvented appliances, such as kitchen ranges and space heaters, is a source of radon in buildings. Scant data exist on radon and its progeny concentrations resulting specifically from the use of natural gas in buildings.[34-35] Concentrations of radon remaining in the gas when it is used in buildings may vary widely as a result of many factors including the wellhead concentration, dilution, removal during processing, transmission time in the distribution line and reservoir storage time which reduce the concentrations by decay.[35]

Liquified Petroleum Gas[35-37]

Before natural gas is distributed to consumers it is processed to remove impurities and heavy hydrocarbons. Some of these hydrocarbons, particularly propane, are recovered and distributed under pressure as liquified petroleum gas (LPG). This process may remove up to 50% of the radon in natural gas,[35,37] but a substantial fraction of the radon will remain in the LPG which is sold by retailers for use in buildings. As in the case of natural gas, the further from the main processing areas, the lower the concentration of radon.[37]

Uranium Mining and Milling Wastes[38-49]

The two most common methods of obtaining uranium in the U.S. are by open-pit and underground mining. Open-pit mining produces the largest amount of waste because of stripping of the overburden from above the ore. For example, in Wyoming in 1974, 2.7 million tons of uranium ore were mined almost exclusively by open-pit, and 107 million tons of waste were generated. In Colorado, 1.2 million tons of ore were mined using underground methods, and only 1.2 million tons of waste were generated.[38] These waste products include solids, liquids and exhaust gases.[38,39] While underground mining produces less waste than strip or open-pit mining, it does expose miners to higher concentrations of radon and its progeny. After the ore is mined, it is sent to mills (usually located in the immediate area) where the uranium is extracted out of the ore, leaving large quantities of waste products called "mill tailings." These mill tailings contain almost all of the radium, thorium and their progeny which were originally present in the ore. Over the lifetime of a typical mill, 40 to 80 hectares (100 to 200

acres) of land may be committed to storing these tailings.[40] As of December, 1977, the magnitude of the tailings exceeded 110 million tons for the active, active standby, and inactive mills in the U.S.[38,40] Radon and radioactive particles containing radon progenitors are released into the atmosphere from the tailings piles as well as during the milling operations (from ore piles, during crushing, grinding and leaching).[38,39] In addition, radon may be dispersed into ground waters and nearby rivers by seepage from liquid waste ponds and by the leaching of tailings piles.

From 1953 to the mid 1960s, the use of uranium mill tailings for construction was common in several Colorado communities, particularly in Grand Junction and Durango. The tailings were used as landfill and backfill, and as aggregate in cement for many homes and commercial buildings. This practice was stopped in 1966 after the discovery that radon and radon-progeny levels inside these buildings were considerably elevated above those found in buildings without tailings. It became apparent that remedial action should be initiated, and guidelines were established by the Surgeon General for permissible radon-progeny levels in these buildings (Table 3).

TABLE 3

Surgeon General guidelines for buildings constructed on or with mill tailings[41].

Indoor WL	Recommendation
>0.05	remedial action indicated
0.01-0.05	remedial action may be suggested
<0.01	no action indicated

A broad survey identified 600 buildings (out of 15,000 surveyed) that have radon-progeny levels above the Surgeon General guidelines and, therefore, require removal of this material. The total cost of the remedial effort, including clean-up of mill tailings piles, will be in excess of $80 million.[42] Congressional concern prompted passage of legislation in the fall of 1978 giving the Nuclear Regulatory Commission authority to license and control mill tailings, and authorizing the Department of Energy to assist States in cleaning up a number of mill tailing sites.[43] The Canadian government has gone through similar experiences.[44]

Phosphate Mining, Milling and Reclaimed Lands[33, 50-64]

The phosphate mining and manufacturing industry as a source of radon is somewhat similar to the uranium mining situation. Phosphate rocks contain naturally occurring quantities of uranium, thorium and radium, which are redistributed among the various products, byproducts and wastes of this industry. The U.S. produces 40% of the total world production of phosphate rock and exports only 30% of the phosphate produced.[50] Over 80% of the total U.S. production occurs in central Florida with the remaining plants located primarily in the west.[51] Most of these plants produce fertilizer, phosphoric acid, and small quantities of elemental phosphorus.[52] Enhancement of natural radon levels occurs during the mining and milling operations, from the use of reclaimed lands, and from the use of various phosphate products and byproducts. These latter sources will be described in subsequent paragraphs.

Most phosphate is mined in an open-pit or strip-mining process. In order to reach the phosphate rock, miners must first remove a top layer called overburden (which ranges from a few decimeters to 12 meters thick) and a second layer known as the leach zone (which can be anywhere from a few decimeters to 1 or 2 meters).[52,53] The ore layer, or matrix, is then exposed and removed, and the top two layers are replaced but not necessarily in the order of occurrence. As a result, material from the leach zone, which has been found to have generally higher concentrations of ^{226}Ra than either the overburden or the ore matrix, is frequently placed at or near the surface[53] which results in increased radioactivity concentrations. Once the ore is removed from the ground, it is stacked, washed, and pumped in slurry form to the beneficiation plant where it is prepared for the mills by upgrading the P_2O_5 percentage. Most western phosphate does not require beneficiation, but it is necessary for

Florida ore. The beneficiation process, which is mainly by physical separation, produces marketable phosphate ore and two separated wastes: slimes which go to a slime pond for settling; and sand tailings which go to a tailings pile.[54] Approximately one ton each of slimes and tailings must be disposed of for each ton of marketable phosphate rock.[33] The rock is now ready for the milling process where it is either mixed with a solution of sulfuric acid and water to produce phosphoric acid and gypsum, or it is placed in an electric furnace along with silica and coke to form elemental phosphorus.[54] The phosphoric acid is further reacted with either ammonia or marketable phosphate rock to form ammonium phosphate or triple superphosphate, respectively, which are utilized in the production of fertilizer.

With respect to exposure to radon and its progeny, there are several problems associated with the mining and milling of phosphate ore and the subsequent redistribution of the naturally-occurring radionuclides. These problems involve the use of water, the accumulation of gypsum piles, and the disposition of reclaimed phosphate lands.

Water used in both the mining and milling processes contains significant quantities of the naturally-occurring radionuclides which are discharged along with the water into large settling ponds. During periods of heavy rains, the ponds, which are often 200 hectares (500 acres) or more, may overflow into nearby streams where the radioactivity is dispersed.

The second problem stems from the fact that phosphate milling involves the accumulation of massive gypsum piles (10 to 30 meters high).[54] This waste gypsum or phosphogypsum is currently stored at mill sites where the natural radionuclides present can be a source of exposure to the general population in much the same way as the uranium mill tailings piles. There is a potential use for this phosphogypsum in building materials, and this will be discussed shortly.

The third problem involves the use of reclaimed phosphate land for residential and commercial development. Over 40,000 hectares (100,000 acres) of land have been mined for phosphate rock in Florida. As of 1977, about 10,000 hectares (25,000 acres have been reclaimed for building purposes, farming and grazing.[51] Since the reclaimed lands are composed of overburden, leach zone material, ore matrix, sand tailings and slimes, they frequently contain ^{226}Ra concentrations of 10 to 30 $pCi \cdot g^{-1}$ which is substantially higher than the 0.1 to 3 $pCi \cdot g^{-1}$ typical of U.S. soils.[29] The increased levels of ^{226}Ra produce considerable quantities of ^{222}Rn which diffuses to the surface and through the foundations of buildings, which leads to a build-up of short-lived ^{222}Rn progeny in the indoor environments.[51] Elevated radioactivity levels on reclaimed lands and ^{222}Rn-progeny levels in buildings built on the land has been extensively investigated over the past few years by the State of Florida and the Environmental Protection Agency (EPA).[51-53, 57-59, 61-62] Recommendations for radiation protection in Florida phosphate lands have also been promulgated by the EPA.[63, 64]

Phosphate Products and Byproducts[33,50,65-68]

The use of phosphate products and byproducts constitutes another source of exposure to radon and its progeny. Two of these products-fertilizers and building materials-will be considered.

As indicated earlier, almost 80% of the phosphate rock mined in the U.S. is used to manufacture fertilizers. The use of phosphate-based fertilizers results in the redistribution of naturally-occurring radionuclides. All fertilizers contain varying quantities of radionuclides, principally members of the ^{238}U and ^{232}Th decay series. Typical radioactivity concentrations in various fertilizer materials made from Florida phosphates were reported by Guimond[50] and range from 1 to 60 $pCi \cdot g^{-1}$. In addition to occupational exposures resulting from the mining and manufacturing of the phosphate products,[55, 56, 59] there are several exposure pathways for the release of radon and radon progenitors to the environment which results from the use of fertilizers.[50, 65] These pathways include: 1) Plant uptake - There is some plant uptake of ^{238}U and ^{226}Ra depending upon solubility, crop, soil and

calcium concentration in the soil. The amount of radionuclide uptake was found to be minor.[50] The escalating use of fertilizers, however, may lead to increases in the radioactivity concentrations found in various crops. 2) Leaching and runoff - It is estimated that over four billion tons of sediment are annually lost to surface waters through erosion. About half of this loss results from agricultural uses, and contains approximately 4.5 million tons of phosphate.[50] Most of this runoff eventually reaches the oceans, but some may collect on river banks and basins where it would be available for plant uptake. Additionally, radionuclides present in the runoff may enter ground and surface water supplies. 3) Fertilizer application - Agricultural workers who apply fertilizer or who work in fields which have been recently fertilized are subject to both direct gamma radiation and to inhalation of radon and radon progeny released to the atmosphere. Considering the total amount of fertilizers applied over the last 80 years, estimates have been made of the exposure that could be received by a typical agricultural worker.[54] Certain states are heavy users of phosphate-based fertilizers[50] and both workers and the general public residing in agricultural areas in these states could receive higher exposures.

Considerable quantities of waste gypsum or phosphogypsum, one of the byproducts of the phosphate industry, are produced each year which must be disposed of in some way (one ton of marketable rock produces 0.6 ton of gypsum).[65] The estimated annual production of phosphogypsum in the U.S. from 1950 to 1973 increased from approximately 1.5 to nearly 24 million metric tons.[66] The ^{226}Ra content of this waste material is typically 30 $pCi \cdot g^{-1}$. The increased production of this waste gypsum has led to a growing interest in developing and using it in building materials. This would solve a number of disposal problems while at the same time creating new environmental ones. This phosphogypsum is being used in building materials in some European countries and Japan[66] although it is not a current practice in the U.S. Florida phosphogypsum was used in the manufacture of wallboard, partition blocks and plaster by at least one company during the eleven year period from 1935 to 1946.[66] The products were mainly distributed in northeastern states. From 1934 to December, 1978, the Tennessee Valley Authority sold waste gypsum and phosphate slag from their fertilizer facilities in Alabama to several concrete block manufacturers in southern states.[67] Annual distributions ranged from 5,000 to 70,000 tons of byproducts. Thousands of buildings in Idaho and Montana have also been reported to have been partly constructed with materials made with phosphate slags.[68]

Building Materials[33, 65, 69-71]

All building materials contain radioactivity that is inherent to the natural raw materials of which they are composed. In addition to building materials containing phosphogypsum, phosphate slag or other atypical higher-than-average concentrations of naturally-occurring radionuclides,[65] even materials of a completely natural origin (such as granite or pumice stone) are sources of radon. Concentrations of naturally-occurring radionuclides, principally ^{232}Th, ^{226}Ra and ^{40}K, in various types of building materials and their effect on indoor radiation levels has been studied in a number of countries. This literature was reviewed in 1975[71] and also summarized in the 1977 UNSCEAR report.[65]

Potable Water Supplies [33, 72-78]

^{226}Ra, ^{222}Rn and their progeny exist in varying concentrations in the groundwater throughout the country. Disequilibrium arising from migration of ^{238}U-series radionuclides via soil water was noted previously. In general, concentrations of ^{226}Ra in common groundwaters are typically 1000 times lower than ^{222}Rn because radon is relatively soluble in water.[73] The solubility of radon in water at atmospheric pressure decreases from 0.5 $L \cdot kg^{-1}$ at 0°C to approximately 0.1 $L \cdot kg^{-1}$ at 60°C.[74] Concentrations of ^{226}Ra may range from trace quantities to over 50 $pCi \cdot L^{-1}$, while ^{222}Rn may be found in concentrations exceeding 50,000 $pCi \cdot L^{-1}$,[33] Distributions of ^{222}Rn concentrations in water in the U.S. and some subregions have been

reported.[75] Population exposure to these radionuclides results from the use of private, commercial, agricultural, public and community groundwater supplies. The primary path of exposure is through ingestion by drinking the water.[75] A secondary exposure pathway is through inhalation of ^{222}Rn progeny. If the water is heated, aerated (such as in a shower head), or agitated (as in a washing machine), the radon is readily released into the atmosphere. Some limited studies have been made of ^{222}Rn-progeny concentrations in homes resulting from the use of tap water.[75-77] This subject is receiving increasing attention[78] since it may involve exposure to large segments of the population. Another possible exposure pathway is through crop uptake[73] which can result if these potable supplies are used for irrigation as they often are in small scale gardens and family farms. This pathway, however, would involve only limited segments of the population.

As indicated, there are a considerable number and variety of both natural and TENR sources of ^{222}Rn and its progeny to which man may be exposed. The primary source to the outdoor environment is by emanation of ^{222}Rn from the earth, but this may be enhanced by contributions from mill tailings piles, phosphate lands, the burning of coal, and other TENR sources. Indoors, the sources include building materials, soil surrounding the building foundation, water, natural gas, and infiltration of outdoor air.[79]

As the radon emanates from soil, water, building materials, etc. and disperses into the air, it is not accompanied by its heavy metal progeny which are relatively immobile. When the ^{222}Rn decays by α-particle emission, its daughter ^{218}Po (Figure 3) is formed as a recoiling positively-charged ion with a range in air of about 50 μm.[80] Initially, the ^{218}Po ion is most likely positively charged because of ionization of the atomic electrons by the emitted α-particle. These ions exhibit a high rate of diffusion in air,[81, 82] are fairly reactive, and interact very rapidly with water vapor, oxygen or other gases to alter their charge state or to form small molecular clusters.[79] Within a very short time, these clusters tend to become attached to aerosol particles suspended in the air (the mean lifetime existence as free unattached ions is of the order of 10 to 50 seconds).[81] The ^{218}Po may be either attached or unattached when it decays to the granddaughter ^{214}Pb. If unattached, the newly created ^{214}Pb undergoes a fate similar to that just described for the initial ^{218}Po. If the ^{218}Po is attached at the time of decay, the ^{214}Pb may, depending upon the size of the aerosol particle, either recoil deeper into the particle or disattach from the particle. This dynamic sequence of events continues throughout the progression of the ^{222}Rn-subseries decay.

The fraction of the activity of the short-lived ^{222}Rn progeny which is absorbed on aerosol particles is called the "attached fraction", and the fraction carried by the molecular clusters (or existing as free ions or atoms) is called the "unattached fraction."[79, 82] This distinction is important since the sizes of the clusters and aerosol particles are considerably different, and therefore do not deposit in the same region of the respiratory tract when inhaled.[79, 83-85] The fraction of unattached

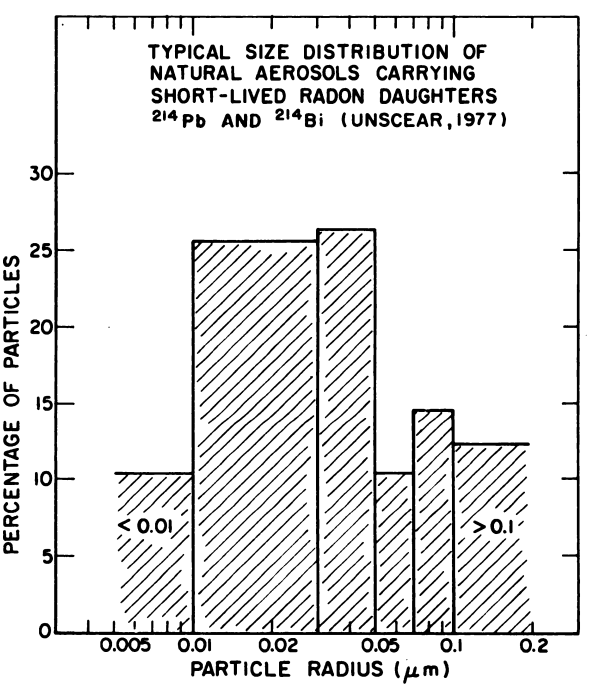

FIGURE 7.

progeny is, however, normally quite small (typically 5 to 10%).[79]

The size and specific gravity of the particles largely determine where the particles will be deposited in the respiratory system and the fate of the particles after deposition.[85] The size distribution of aerosol particles carrying ^{222}Rn progeny is dependent on the size distribution of the natural aerosol which is present. This size distribution is relatively constant[79] in both outdoor air and in the ambient atmosphere within typical buildings. The typical size distribution of aerosol particles carrying the short-lived ^{222}Rn progeny ^{214}Pb and ^{214}Bi is shown in Figure 7.[79, 86] As indicated, nearly 80% of the particles carrying the ^{214}Pb and ^{214}Bi have radii between 0.01 and 0.1 μm. These are mainly deposited in the pulmonary region,[83-85] in contrast to the progeny in the unattached fraction which are deposited in the upper respiratory tract. The implications of these particle size considerations in determining lung dose and the impact on respiratory cancer, particularly in uranium miners, have been discussed in a large number of studies.[83, 84, 87-91]

In assessing the potential radiation hazard due to ^{222}Rn and its progeny, the important factors include the degree of radioactive equilibrium between ^{222}Rn and its progeny, the relative attached and unattached fractions, and the aerosol particle size distributions. Each is time dependent and is influenced by a large number of variables. The levels of ^{222}Rn and its progeny within buildings and their dependence on both locally-dependent and time-dependent variables have been extensively studied by many investigators.[12, 16, 58, 80, 84, 92-108] Two of the most important variables are the ventilation rate[58, 84, 96, 97] and the plateout of the aerosol particles by deposition and adsorption on walls and surfaces.[90, 105, 107] Increased ventilation or high air change rates usually decreases aerosol particle concentrations within buildings, and at the same time decreases the residence time or "age" of the air. The net result is a greater degree of radioactive <u>dis</u>equilibrium, an increase in the unattached fraction, a decrease in the WL, and a reduction in the dose to the lung. Source dependent or locally dependent factors[99], such as the type of building materials and the location of the building or rooms inside the building, also influence the levels of radon and its progeny in the room. The dependence of radon emanation rates on meteorological parameters was noted previously. These parameters, such as barometric pressure, wind speed, air and soil temperature, relative humidity, etc., similarly affect the concentrations of ^{222}Rn progeny in buildings.[92, 94, 98, 99, 103] Steinhäusler[99] evaluated the dependence on 24 meteorological variables, and found great daily and seasonal fluctuations in indoor radionuclide concentrations even under constant ventilation conditions. In general, because of the strong influence of these time dependent variables (e.g., the ventilation rate or meteorological effects), it is presently impossible to correlate the levels of radon and its progeny in buildings to the known radon sources or the locally dependent factors (e.g., the radium content of the building materials). As evidenced by just the titles of a number of studies that have been conducted in the past few years (cf., references 94-108), the effects of many of these variables have been studied either individually or several at a time. No detailed models exist, however, which can adequately predict the levels of radon and its progeny for given conditions or simultaneously treat all of the antecedent-consequent relationships that are involved.

This review has attempted to demonstrate that the sources and behavior of radon and its progeny are exceedingly complex. Inasmuch as "radon" may be the most serious problem we have in environmental radiation protection,[109] greater understanding and more complete solutions to this problem are urgently needed.

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ADDENDUM

Despite the seemingly excessive number of references cited above, the author wishes to note that it is not by any means an exhaustive list. Hundreds of additional sources may be found in some of the reviews cited herein. The extensiveness of the literature is such that Harley[26], a few years ago, noted that a bibliography of a thousand references to radon and its progeny can be easily assembled. The present list has barely touched upon the vast biological, health effects and epidemological work, and has excluded the additionally extensive literature relating to various measurement methods.

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THE BIOLOGICAL AND HEALTH EFFECTS OF RADON: A REVIEW[+]

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Human knowledge of radon began near the turn of this century when the discovery of radioactivity followed closely the discovery of X-rays. Thus began an exciting period in the fields of physics, chemistry and biology. Radium especially became important when its intense gamma radiation was found to be as effective as the X-ray in its interaction with, and damage to, biological systems. Radium could be sealed into a metal tube or needle and surgically implanted in living tissue to provide in situ irradiation of various cancers, whereas X-rays were directed to the cancerous growths from outside the body and were thus required to traverse normal tissue surrounding the growth. Medicine had acquired a new surgical tool. The physicists, in developing the decay schemes of the radioactive materials, found that it was not radium but rather one of its decay products which provided the intense gamma radiation. Radon, a gas and first decay product of radium, was parent to the needed gamma radiation and for the same intensity radon sources or seeds could be much smaller than the radium needle. Also the long half life of radium, while it permitted a constant dose rate, required that the needle be removed after treatment, whereas radon, with a short half life and consequent rapidly decreasing dose rate, could be contained in glass seeds and left in situ in many applications. In addition, radon became useful to the medical profession which, with its suppliers, promoted the use of radon for the common good by developing ointments and salves containing radon for topical application. Other entrepreneurs arranged to let people bathe in radon water, inhale radon, drink water which contained radon, and intravenously inject radon-containing solutions, all to promote the general good health of the people.

The physicists had been studying the decay schemes of uranium and much information was published in the scientific journals of the day. A first book on radioactivity appeared in 1904[1] and was followed over the next two decades by many others from many countries. The extensive treatment by Rutherford, Chadwick and Ellis appeared in 1930[2]. Jennings and Russ[3] have provided a description of the production and use of radon seeds.

The chemists had been busy developing the field of radiochemistry and, of interest here, there was the production of emanation sources (another name for radon, more specifically radium emanation). It was desired to have sources which could release radon continuously and this was accomplished by incorporating radium into colloids or organic structures. These sources could release nearly all the radon formed without losing radium and were essentially dry sources, therefore handy and useful. The radiochemistry of the period, as well as the production of emanation sources, has been described by Hahn[4].

The biologists began to study the toxic nature of radium, radon and X-rays soon after their discoveries and reports of small animal experiments appeared as early as 1904[5-7]. Thus began the studies of the biological effects of radon. Experiments with the X-ray and the gamma emissions from radon were designed to demonstrate the utility of the radiations in the treatment of maligant diseases, but many studies with radon were directed to the discovery and promotion of the healing powers of radon therapy. Much of the work with radium and radon was summarized by London[8].

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In 1879 Harting and Hesse[9] had described as lung cancer the illness miners contracted in the uranium mines of Schneeberg, Germany, and in those of nearby Joachimsthal, Czechoslovakia. Mortality from lung cancer was near 75 per cent, with the incidence greater for miners than for the masons and carpenters working in the mines. In 1913, Arnstein[10] reported from a new study that the lung cancer was a squamous-cell carcinoma, and in 1924 Ludewig and Lorenser[11], familiar with the carcinogenic action of X and gamma radiation, suggested that the lung cancers of the Schneeberg miners, who were now mining the uranium ore for its valuable radium content, might be caused by the radon in the mines. The radon concentrations in these mines averaged about 4×10^{-9} curie per liter, compared to levels of about 10^{-12} curie per liter far away from the mines.

Thus did radon become suspect as the cause of lung cancer in uranium miners, and from that time to the present much work has gone into studies of its carcinogenic activity as well as determinations of maximum recommended concentrations. Radon still had value as a therapeutic agent through use of radon seeds, and emanation therapy continued. The ubiquitous nature of radon was known but lung cancer was considered to be an occupational problem. Many reviews of the mine data and animal experiments are available [12-15].

Not everyone agreed that radon alone caused the lung cancer in the miners, and Lorenz[12], after reviewing the extensive literature on the subject, concluded that radon could not be the sole cause of the lung cancer in uranium miners. Lorenz mentions as contributing factors pneumoconiosis from dust in the mines, chronic irritation caused by respiratory diseases, arsenic, other radioactive substances, and perhaps a hereditary susceptibility.

Many experiments have been conducted to elicit the biological effects of radon inhalation. Most of these were acute-lethal and subacute lethal exposures so that effects might occur in reasonable periods after exposure and they had obvious ill effects on the health of the animals. Some longterm exposures did not appear to permanently affect the health of the animals despite severe transient effects. Morken has summarized most experiments and provided confirmation of the many of them as well as new information[13,16,17,19,20] and has provided new information on long term expsoures[20,21]. Scott has reported on the histopathology of mice exposed to radon, confirming much previous work and offering new information[23].

In general, sub-lethal exposures to radon produced effects on life shortening and body weight similar to those found with X or gamma radiation. Two specific differences did appear: red blood cell forming tissue was only slightly, but permanently, injured whereas the other blood cell forming tissues responded in the expected manner, and the growth pattern was altered and produced permanent stunting. Spleen evidenced the greatest early damage but also showed rapid repair, and late injury appeared in the kidney. Lung and trachea showed no lesions early after exposure but the epithelial lining of the small bronchi showed changes at 2 months, with definite injury at 7 months. Of all the effects, only the red cell depression was not proportional to dose.

In these experiments the tissue-absorbed dose was calculated from the measured activities and weights of tissues, a standard practice following the definition of absorbed dose. Whole body doses ranged from 50 to 300 rad. Although radon was shown to be toxic and could provide the kind of lesion which was thought to precede lung cancer, early experiments were generally unsuccessful in producing lung cancer. Most of these experiments used an exposure system in which only the radon was inhaled, with the decay products of radon in air having been effectively removed either deliberately or inadvertantly from the inhaled atmosphere. With the recognition and confirmation[24-26] that radon in air which contained dust produced a radioactive aerosol which could be inhaled along with the radon, the problem of the miner's lung cancer took a new turn.

Radon-222, a noble gas and decay product of radium-226, decays with a half-life of 3.82 days and the emission of an alpha particle to radium-A (RaA, Po-218), which in turn decays with a half-

life of 3.05 minutes and the emission of an alpha particle to RaB (Pb-214). RaB decays with a half-life of 26.8 minutes and the emission of a beta particle and coincident gamma ray to RaC (Bi-214). RaC decays with a half-life of 19.7 minutes primarily by beta particle emmision and coincident gamma ray to RaC' (Po-214) which decays almost instantly to RaD (Pb-210) by the emission of an alpha particle (a few RaC atoms take an alternate path through RaC'', Tl-210). The long half-life of RaD, 22 years, removes consideration of the remainder of the decay chain from the immediate problem; following RaD are RaE (Bi-210), RaF (Po-210) and finally RaG (Pb-206) a stable isotope. The products RaA through RaC' have historically been called the "active deposit" of radon because of their propensity as metallic ions to attach to any nearby surface. In normal atmosphere these products are found attached to dust particles, most frequently perhaps because the first-formed RaA will so attach. For the brief time from formation to attachment the RaA atom will exist in the unattached state and any specified volume of dust will show both attached and unattached atoms. Attached atoms will decay through the chain to produce attached RaB, RaC and RaC'. It is a consequence of the decay sequence that if the process is allowed to progress for a long time (about 4 hours for the radon series) the radioactivity of each of the isotopes will equal that of the parent radon. If competing surfaces such as walls are provided much of the active deposit may be found on those surfaces, to effectively remove them from the air.

The situation in the mines favored the attachment of the decay products to dust particles. Some of these particles when inhaled will deposit in the lung. Then in addition to the dose from inhaled radon, there will be a dose delivered from the radioactivity carried on the dust particles. Indeed, the dose from radon now waned in comparison to that from the radioactive aerosol.

The deposition of dust particles in the lung has been well studied and well reported (e.g.[27]). It is necessary to understand the morphology and respiratory physiology of the lung as well as the fluid dynamics of aerosol particles to permit a proper evaluation of the dose delivered to the lung from the decay products of radon. The lung is a complex organ mechanically, but may be divided into two major parts below the larynx for consideration of dust particle deposition. Inhaled air and dust are carried through a series of branching airways, the bronchial tree, until they reach the alveolar region where the usual gas exchange occurs. Dust is deposited along the length of the bronchial tree and in the alveolar region, with particle size separation occurring according to air flows, particle parameters of inertia and diffusion, and the branching. Larger particles deposit primarily in the upper parts of the bronchial tree and smaller particles in the deeper portions of the lung. Deposition is not total and the fraction of a given size particle which deposits in each portion of the lung has been well studied[27]. The unattached ions are almost completely removed in the region above the larynx if breathing is through the nose, but for mouth breathing these ions are likely to be deposited in the trachea and major bronchi.

Solid materials which deposit in the lung undergo removal, sometimes incomplete, by several mechanisms. The primary clearance mode in the bronchial tree is the ciliated epithelium which carries particles to the mouth and this method is rapid, total clearance being accomplished in an hour or less. Clearance from the alveolar region is less rapid, principally by dissolution for soluble dusts, a relatively rapid process, and by phagocytosis, a slow process, but other methods are thought active. Dusts can remain in the lung for long periods. The slow removal from the alveolar region, important for consideration in the dust-caused diseases of the lung, does not play a significant role because of the relatively rapid decay of the decay products of radon which may have been deposited.

The Schneeberg lung cancer, bronchogenic in nature, appeared to occur about one-third the way down the bronchial tree from the trachea. It has been a fairly simple matter to show, when suitable parameters are chosen, that the greatest dose to bronchial tissue might occur in the region of the Schneeberg lung cancer. The rapid transport of

the radioactive dust toward the trachea, the relatively short life of the radioactivity, the selectivity to particle size, the geometry of the tissue, and the ventilation rate are all considered in the calculation of the dose. The geometry of the situation involves consideration of a thick-walled tube with the activity on its inner surface. Because only alpha particle energy applies, the penetration of the alpha particle into the wall of the tube probably cannot exceed the range of the alpha particle in tissue; about 45 μm for RaA and 70 μm for RaC'. Limiting the thickness in this way results in a smaller mass in which the energy is released and promptly results in a greater dose than would otherwise be calculated; this method of calculation is justified only by the geometry involved but will result in improper comparisons with doses calculated in the normal way.

More recently, attention has focussed on a target method, in which the dose absorbed in small volume, commonly one cubic micrometer, at the presumed depth of a cell nucleus is calculated for the radiation from a particle in transit on the inner surface of the tube. From the methods proposed it appears that the dosimetry is arranged to maximize the dose in the region of the Schneeberg lung cancer.

Parker has discussed the possibilities and probablities, the failures and the successes of these various methods in light of the great biological variations which occur[28]. Walsh[29] has further reviewed these methods and others continue to improve them[30,31] with the intent to provide a firmer basis for a regulatory level for uranium mines.

Application of the methods of dosimetry to environmental situations requires changes in some parameters. Lung morphometry and physiology will remain the same, but the breathing rate, which brings in the dust, must change to allow for work, play and rest periods. Natural environmental particle sizes are generally smaller, of a size to produce greatest deposition in the alveolar region.

Hofmann et al[32] have estimated dose to the lung for various age groups and found a strong influence of age and physical activity and a slight dependence on sex and weight. Maximum dose from natural radioactivity appeared in the 5-to-9 years age group. Adult doses were received chiefly at places of employment.

In the 1940's the maximum permissible concentration of radon in air for continued human exposure was set at 10^{-11} curie per liter in equilibrium with the short-lived decay products, as recommended by a consideration of the radon exhaled from a known body burden of radium and including a factor of safety of 100[34]. In the 1950's this was raised to 10^{-10} curie per liter in equilibrium with the short-lived decay products. With consideration of the decay products only this latter level was changed to include just the alpha particle energy potentially available in one liter of air from any combination of the decay products, and was designated the Working Level (WL). This WL represented a concentration or dose rate, and exposure to 1 WL for a 168-hour work week constituted a dose for 1 Working Level Month (WLM). Reference[15] describes how these regulatory levels were derived. The annual permitted exposure was then 12 WLM, since reduced to 3 WLM. The WL is a useful air-equivalent unit of dose rate because it contains a measure of the energy available for release in the lung and is not subject to the vagaries of the lung parameters; it also does not require equilibrium of the decay products, as did the radon levels. Radioactive equilibrium rarely occurs except in a stagnant situation and even then the presence of surfaces, such as walls, removes the decay products from the air. Most mine and environmental atmospheres contain decreasing isotope radioactivities for each isotope which occurs further down the chain from radon.

Epidemiologic studies of uranium miners in this country have used the WLM as the dose unit, and these studies have been well reported[35,36]. While the evidence certainly indicates lung cancer incidence among uranium miners is greater than for non-miners, there are other possible agents in the mines. Also reported[37,40] are non-uranium mines in which the radioactivity levels are elevated above background values and sometimes equal

to those in uranium mines, but with no corresponding high incidence of lung cancer such as afflicts the uranium miner. A more recent report[38] suggests that despite strict controls on mine atmospheres for the past twenty or more years the high incidence of cancer has not abated, and a non-malignant respiratory disease in miners, thought due to diffuse parenchymal radiation damage, is approaching cancer in importance as a cause of death. Current dosimetry methods do not apply to other than the bronchogenic cancer.

Experiments with radon and dusty atmospheres have been reviewed[22]. These have been unsuccessful in producing lung cancer when the natural aerosol was used, but radon inhalation during or after exposure to a noxious dust has produced tumors and cancer, mostly pulmonary, rarely bronchogenic. It appears that once a lesion has been produced by chemical agents radon inhalation enhances the effect; radon inhalation followed by application of the noxious dust does not provide enhancement. Recalling that Lorenz[12] anticipated this result, current dosimetry concepts might not relate to present concerns with radon in environmental atmospheres.

Radon itself has not completely disappeared from the scene. Radon therapy is still being studied and applied for its health-saving properties[41,42].

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TECHNIQUES FOR MEASURING RADON IN BUILDINGS

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INTRODUCTION

Present techniques for monitoring radon and radon daughters, although not in all cases the most convenient or efficient that might be desired, are adequate for almost any requirement. Probably the most critical, present deficiency is the lack of commercial versions of some of the newer instruments. Much of current radon monitoring is being done with instruments that are either home-made or have been obtained by special arrangements.

There is an abundance of available techniques, each one having attributes that make it suitable to certain applications and it is necessary to be selective in satisfying the objectives of a particular mission. By confining attention to buildings, a few methods are eliminated from consideration but a formidable number remains. In the following presentation, some of the more practical methods will be identified.

Before discussing specific methods, it may be useful to touch on accuracy and precision, important considerations in connection with all measurements. The following generalization applies to most any kind of radon monitoring: errors of accuracy and precision of individual measurements are small and generally insignificant compared to errors in applying and interpreting the measurements. Like other air pollutants, radon concentration varies greatly with time and location although probably to a greater degree. The errors of most radon measuring techniques are acceptably small but it is of questionable utility to measure a particular concentration with an accuracy, say, of ±10%, recognizing that an hour later the concentration at the same location may differ by a factor of two or that the concentration in an adjoining room of a building differs substantially from the one that has been measured. Actually, radon variation is more likely to be 10-fold than 2-fold over the period of a day or longer. Generally speaking, single measurements are not very useful and one must resort to multiple measurements, either by manual or automatic means, to develop complete information.

This assertion about small instrumental errors is predicated on proper calibration. In this regard, it is vital to calibrate instruments and to intercalibrate with other groups periodically. Moreover, it is inadvisable to accept the calibration of a radon instrument as received from a commercial supplier. Occasionally, commercial instruments are brought to our laboratory by purchasers who are not obtaining correct measurements. In every instance to date, the instrument itself has been of good quality but simply was not calibrated properly at the factory. The problem usually can be corrected with a few hours of effort.

To present methods in some semblance of order, it's convenient to separate them by applications. If the mission under consideration is monitoring, one is concerned mainly with the air concentration of either radon or radon daughters or both. If there is a requirement to calculate lung doses to building occupants, additional information may be needed, adding a degree of complexity to instrumental requirements. For example, radon daughter ratios, fraction of uncombined radon daughters, and particle size distribution enter into these calculations. Taking another direction, if measurements are to be diagnostic, sources of radon and rates of radon input may be important, thus necessitating measurements of radon emanation from building surfaces and, possibly, radon concentration in water supplies which can be a substantial, even a dominant source of air radon in buildings. And finally, since air concentration is a function of ventilation rate as well as radon input rate, the rate of air exchange is a measurement of possible interest.

AIR CONCENTRATION

Measurements of air concentration can be sub-

divided into three groups: prompt (otherwise known as instantaneous or grab sampling methods) methods in which an air sample is obtained either instantaneously or over a brief sampling period of 5 to 10 minutes; continuous readout monitors that continuously sense and register the concentration; and time-integrating monitors that yield a single, average concentration for an extended time period from a few days to a week or more.

Prompt Radon

Scintillation flask. The scintillation flask,[1,2] popularly known as the Lucas flask, is one of the oldest, most reliable, and widely used methods both in the laboratory and in the field. Required equipment consists of one or more scintillation flasks and a photocathode counter, both available from commercial sources.

The scintillation flask is a small glass or plastic vessel equipped with one or two valves and lined with a phosphor (silver-activated zinc sulfide) which emits light flashes when bombarded with alpha particles. A sample of whole air is collected in the flask which is then counted with the photocathode, usually after a delay of a few hours to allow radon daughters to equilibrate with the radon. In routine use, its sensitivity is about 1 $pCi \cdot L^{-1}$ but with care and long counting periods, a sensitivity of <0.1 $pCi \cdot L^{-1}$ can be attained. Portable, battery-powered photocathode counters are available which enable the measurements to be performed entirely in the field.

Two-filter method. The two-filter method,[3] a later development than the scintillation flask, is intended primarily for the field. It requires a two-filter tube, which is not available commercially, in addition to an air sampling pump and a portable alpha counter, both of which can be obtained commercially.

The two-filter tube is a metal cylinder with a high efficiency filter at each end. Sample air is drawn through the tube for five or ten minutes. The front filter removes all particulates including radon daughters from the air stream allowing only radon to enter the tube with the air. While in-flight through the tube, some of the radon decays to radium-A, part of which is collected on the downstream filter, the rest diffusing to the wall of the cylinder. Immediately after sampling, the downstream filter is removed and alpha-counted for several minutes. Radon concentration can be calculated by a simple formula that takes into account tube dimensions and sampling and counting times.

The entire measurement is completed in the field in a total time of 15 to 20 minutes and measurements can be repeated indefinitely with the same apparatus. Sensitivity is dependent on tube volume. Concentrations on the order of 5-10 $pCi \cdot L^{-1}$ can be measured with a 1 liter tube; sensitivities as low as 0.1 $pCi \cdot L^{-1}$ can be attained with tubes of much larger volume.

Pulse-type ionization chamber. Measurement of radon by the pulse-type ionization chamber[4] has been the standard method at EML for 30 years. Its application in the field is somewhat less convenient than other methods and so it is seldom used in the field unless extreme sensitivity is needed. However, it serves as a standard for calibrating other radon instruments. It is not made commercially.

A whole air sample is collected in a one-liter glass flask and then returned to the laboratory for analysis in a pulse-ionization chamber. A separate flask, protected by a padded shipping container, is required for each measurement rendering the method awkward if more than two or three measurements are required. The method is very accurate and has a sensitivity of about 0.01 $pCi \cdot L^{-1}$.

Continuous Radon

Because of the dynamic behavior of radon in air, it is rarely sufficient to obtain a single grab sample. Any of the foregoing methods can be repeated periodically to cover a longer time span but it is far more convenient to use a monitoring instrument specifically designed for the purpose although considerably increased instrument size, complexity, and cost are necessary to attain the added convenience.

Three kinds of continuously-reading radon

monitors have been developed to serve this purpose: the continuous flask monitor; automated two-filter monitor, and a diffusion electrostatic monitor.

Continuous flask monitor. This instrument consists of a scintillation flask permanently mounted in a photocathode counter. Air is drawn continuously through the flask so that the counter tracks changes in radon concentration as they occur. The first instrument of this type was designed at the former HASL in the mid-50s[5] and similar instruments have been reported several times since by other groups. A commercial version has become available.

The fallacy in this arrangement is that even if the inlet air stream is filtered, radon daughters remain in the flask long after the radon from which they originated has passed through. Since the daughters continue to emit alpha radiation, they mask the changes in concentration which the instrument is designed to monitor. This problem persisted until two years ago when an experimental and theoretical procedure was developed at EML[6] to correct for the previously-deposited radon daughters. This procedure enables one to calculate the correct radon concentration in each measurement period by taking account of alpha activity that was registered in preceding measurement periods. Therefore, the continuous flask monitor is now a reliable instrument that is capable of detecting varying radon concentrations for an indefinite length of time. The approximate sensitivity is 0.5 $pC \cdot L^{-1}$. The instrumentation is not truly portable but can be moved and set up readily by one person.

Automated two-filter radon monitor.[7] Basically this is a two-filter tube with a scintillation-photocathode detector positioned in front of the collecting surface of the second filter. Air is pumped through the tube continuously and the detector continuously counts the accumulated radon daughters. Obviously, this technique also is vulnerable to interference from previously deposited radon daughters. But the interference either can be eliminated by providing a filter transport mechanism to replace the downstream filter periodically or it can be corrected mathematically by an equation[7] that takes the previously collected activity into account. Therefore, this instrument also is capable of recording radon concentration at selected intervals for an indefinite period. The lower limit of detection is inversely proportional to the volume of the tube and sensitivities of 0.01 $pCi \cdot L^{-1}$ have been achieved. However, the sensitivity of a unit that can be moved readily is on the order of a few tenths of a picocurie per liter.

Diffusion-electrostatic radon monitor.[8] This monitor uses a different sampling principle but the method of detection is similar to the monitors just described. The sensitive volume is a 1-liter hemisphere made of a wire screen covered by a light-tight porous foam. At the center of the hemisphere are a scintillator and photocathode detector. A high potential is applied between the wire screen and a disc of aluminized Mylar covering the scintillator. Instruments of this type have not been produced commercially.

In operation, ambient air enters the hemisphere through the porous foam by molecular diffusion. Within the hemisphere, radon decays to radium-A which, being positively charged, is drawn to the aluminized mylar cathode where it deposits and decays to radium-B and radium-C. The alpha decays are detected by the photocathode counter and recorded. The concentrations within the hemisphere follow the ambient radon concentration after a time lag. Therefore, in principle, the monitor is continuously detecting the ambient radon concentration. But, as in the case of the other continuous radon monitors, the changes in ambient radon concentration are masked by the previously deposited radon daughters. Probably, a procedure could be devised by which this phenomenon could be corrected but it has not been done and, at the moment, this effect compromises the utility of the instrument. Another deficiency is a pronounced humidity dependence of the response to radon concentration.[9]

Time-Integrating Radon Monitor

Another class of instrument that measures radon concentration over long periods of time is the time-integrating monitor. This class represents an improvement in simplicity and a lower cost with respect to the continuously-reading

radon monitor. In monitoring radon for compliance with regulations or to assess biological hazard, average concentration is the most useful measurement and time-integrating systems were developed specifically for that purpose. The first and probably most widely used time-integrating method for radon was developed by Sill[10] for studies of radon distribution around tailings piles. Field equipment consists of a 40-liter Mylar bag and an aquarium pump powered with a storage battery. A whole-air sample is collected over two days by pumping air into the bag at a very slow rate, 10 milliliter per minute. Analysis of radon is performed in the laboratory by discharging the sample from the Mylar bag through a cold charcoal trap and then transferring the radon from the charcoal to a Lucas flask for counting. The result is a 48-hour average radon concentration. The method is very sensitive, capable of measuring concentrations as low as 0.01 $pCi \cdot L^{-1}$, and it has been used with success for many years. Commercial equipment is available.

A more recent instrument, a passive environmental radon monitor,[11] was developed at EML for its own ambient radon studies. Its principle of measurement is similar to the diffusion-electrostatic monitor in that it samples by molecular diffusion and collects radium-A ions in an electrostatic field. However, instead of a photocathode counter, detection is by means of a thermoluminescent lithium fluoride chip which continuously absorbs and stores alpha decay energy for as long as the monitor is operating. After a designated monitoring period, usually one to two weeks, the chip is removed and analyzed in the laboratory. Since there are no electronic circuits, the cost of the monitor is very low. Moreover, there is no power consumption, the electrostatic field being provided by dry-cell batteries. Therefore the monitor can be placed anywhere and operated for an indefinite period of time. Sensitivity is about 0.03 $pCi \cdot L^{-1}$ in a one week measurement. A commercial instrument of this general type has been advertised but it is much less sensitive than the EML design and is humidity dependent.

Prompt Radon Daughters

Methods are available to measure radon daughters either in terms of working level or in terms of the individual concentrations of RaA, RaB and RaC.

Kusnetz method. The most practical and popular method for measuring working level was developed by Howard Kusnetz[12] for uranium mines in the mid 1950s. Apparatus consists of a portable air filter sampler and a portable alpha counter, both of which are commercial instruments. Air is sampled through a small-diameter filter for five minutes and then after a delay of from 40 to 90 minutes, the filter is counted for gross alpha activity. Working level is calculated by a formula involving the count rate, sample volume, and a correction factor for decay. In the original method, the alpha activity on the filter was measured with a simple ratemeter but as radon daughter concentrations in mines later were reduced by increased ventilation, it became necessary to substitute a scaler for the ratemeter to achieve better sensitivity. Scalers are used almost exclusively today and concentrations as low as 0.0005 WL can be measured with little difficulty.

Several variations in the basic method have appeared since Kusnetz introduced it, the one attracting the most attention being described by Rolle.[13] The main difference in Rolle's method is that the alpha count is made after a delay of only 5 to 10 minutes after sampling.

Tsivoglou method. The individual concentrations of radon daughters can be measured by either the Tsivoglou method or by alpha spectrometry. Tsivoglou,[14] a colleague of Kusnetz, also developed his method for use in uranium mines in the days when concentrations were orders of magnitude higher than today. The method uses identical equipment. The sampling procedure also is the same but the collected alpha activity is measured three times at prescribed intervals in the 30 minutes immediately after sampling. From the three counts, the concentrations of RaA, RaB, and RaC, can be calculated with three equations and, of course, working level also be calculated.

Like the original Kusnetz method, the original Tsivoglou method used a ratemeter but sensitivity is improved tremendously by substituting a scaler. In the modified Tsivoglou method introduced by Thomas[15] about 8 years ago, the alpha activity is measured by means of a scaler in three intervals over 30 minutes. This method is adequate for concentrations on the order of a few tenths of a picocurie per liter.

Another variation on the Tsivoglou method has been described by Raabe and Wrenn.[16] Again, sampling is the same as in the Kusnetz method but alpha counts are made repeatedly at closely-spaced intervals beginning immediately after sampling and continuing for almost any desired period from 15 to 20 minutes to an hour or more. Calculation of radon daughter concentrations involves fitting the resultant alpha decay curve by a least-squares technique.

Alpha spectrometry. Still another variation of the Tsivoglou method employs alpha spectrometry to count RaA and RaC' separately.[17] Sampling equipment and procedure remain the same as in the previous method but filter counting is performed with a two-channel alpha analyzer. Usually two counts are made in the first 20 minutes after collection.

Conceptually, this method affords better precision than methods that rely on gross alpha counting but in practice, the air sample is sharply limited compared to the other methods because solid state detectors, which are used for alpha spectrometry, are very small. Hence, the methods based on total alpha counting, not being limited in sample volume, are capable of much better sensitivity.

Working level meter. Finally, there is a class of instruments popularly known as working level meters.[18,19,20] Several measure working level only whereas one type measures the individual concentrations of RaA, RaB, and RaC. The first type of instrument is available commercially; the second is not. These instruments more-or-less automatically perform the measurement and register a result in a span of 5 to 10 minutes.

The original versions, developed for use in uranium mines, were not sensitive enough for measurements at natural levels above ground; both types of instrument are now available in models that could be used in buildings.

Continuous Radon Daughters

Proposed methods for measuring radon daughters continuously have not yet been shown to be reliable. Fixed filter samplers have been developed with either alpha or beta detectors that continuously count the deposited radon daughters but the problem of previously-deposited radon daughters, mentioned in connection with continuous radon monitors, has not been solved.

Time-Integrating Radon Daughter Monitor

There are two techniques which have been developed for time-integrated measurements of radon daughter concentration in air. Both give results in terms of working level. One method has been shown to be reliable and practical over many years of use whereas the other, although offering promise, has not yet been perfected for monitoring at low concentration.

The first method, developed by Schiager at Colorado State University,[21] uses a thermoluminescent (TLD) detector in an active air sampling system. The monitor consists essentially of a membrane filter and holder containing a TLD and a low-volume air pump. Air is drawn through the filter continuously at 100 milliliter per minute for a total sampling period of about one week. The TLD is positioned in front of the filter where it absorbs and stores alpha decay energy. At the end of sampling, the TLD is removed and analyzed in the laboratory.

Instruments of this type have been used extensively in Grand Junction, in investigations of tailings piles, and various other situations. We have used our own version[22] in studies of ambient radon daughters in buildings and in outside air. Sensitivity is about 0.0005 WL in 168 hours. Commercial versions have not been produced.

The other system uses track-etch film in a completely passive mode of measurement.[23] One centimeter square pieces of cellulose nitrate film are suspended for a long period of time in the air to be monitored. An alpha particle, upon striking the film, damages the surface. Each point of impact can be enlarged by etching the film in an alkaline solution. The resultant pits are counted with a microscope.

The potential advantages of track-etch film are low cost, simplicity, and the feasibility of extensive coverage, if needed. So far it has not been widely used because of inaccuracy and poor sensitivity. For example, year-long exposures are needed to obtain sufficient track densities at concentrations on the order of 0.01 WL. At that concentration, the precision of measurement is about ±50%.

UNCOMBINED FRACTION AND PARTICLE SIZE OF RADON DAUGHTERS

Measurements of uncombined fractions of radon daughters and of the particle size distributions of radon daughters are obtained by prompt sampling methods.

Uncombined radon daughters, which are single atoms or molecules in air, are measured by diffusion techniques. We have used the diffusion cell,[24] developed by Mercer at the University of Rochester, and the wire screen method,[25] developed by George at EML, with equal success. In both methods, two measurements are made simultaneously, one of the uncombined radon daughters alone and the other of total radon daughters. The ratio of the two measurements is the uncombined fraction. Both the diffusion cell and the wire screen collect uncombined daughters with good efficiency while allowing combined daughters to penetrate freely. The collection efficiency for uncombined daughters must be determined experimentally and typically is in the range of 60 to 90% for a screen of 60 mesh size. The collection efficiency need not be very high but it must be reproducible.

Total daughter concentration is obtained by conventional filter sampling. Immediately after sampling, both the filter sample and either the diffusion cell or wire screen, depending on which is used, are alpha-counted by either the modified Tsivoglou or the Raabe-Wrenn method.

Neither the Mercer cell nor the wire screen device are available commercially but the wire screen device can be assembled readily from conventional air sampling components. The Mercer cell must be machined to rigid specifications.

Radon daughter particle sizes are most readily measured with diffusion batteries.[26] Several different types of diffusion battery that are suitable for measuring radon daughters have been developed but none is available commercially.

In passing through a diffusion battery, a portion of the radon daughters deposits on the battery walls in an amount that is inversely related to particle size. Thus, the smallest particles penetrate least through the battery whereas large particles penetrate much more freely. Collection efficiency as a function of particle size can be calculated accurately by means of diffusion equations. By measuring the penetration of radon daughters simultaneously through several batteries each with a different collection efficiency, a rough size distribution can be determined. Typically we use three batteries and a reference filter, four air pumps, and four alpha counters.

RADON EMANATION

Two methods are in common use for measuring radon flux from soil and from building surfaces, the accumulator method and the charcoal canister method.

The accumulator method,[27] also known as the flux can method, has been in use for 20 or 30 years and enjoys universal acceptance. A can of nominal size (20-100 liters), open at one end, is placed open-side-down on the emanating surface and sealed. After a brief period, usually no more than 30 minutes, radon which has collected in the can is sampled through a valve with a scintillation flask and measured. Radon flux can be calculated from the increase in radon concentration, the time of collection, and the dimensions of the can.

The can, itself, can be constructed very easily; the method is simple, reliable, and quite sensitive. Since the measurement is performed

in a brief period, it is analogous to the prompt sampling methods for air concentration.

In the charcoal canister method,[28] a fairly recent development, a canister from a conventional personal respirator is modified by removing the metal casing from one face. In making a measurement, the canister is placed, open-side-down, on the emanating surface, sealed, and left in place for 1 to 2 days. Emanating radon is absorbed completely in the charcoal. After collection, the canister is analyzed in a gamma spectrometer for the radon daughters which have grown to equilibrium with the radon.

This method is simple, sensitive, and reliable. It is easy to apply in the field and because of the low cost and convenient size of the canisters, many measurements can be obtained simultaneously with little effort. Since the collecting period is one day or longer, the method is analogous to the time-integrating methods for air concentration.

RADON IN WATER

The simplest way to measure the concentration of radon in water is to collect the water in a plastic container and count the radon daughter activity in a gamma spectrometer. This technique was reported 15 years ago by Lucas.[29] Using a conventional 1 quart polyethylene bottle, he obtained a sensitivity of about 100 $pCi \cdot L^{-1}$.

The sensitivity of Lucas' method can be improved by using a modified Marinelli beaker for sample collection and counting.[30] The Marinelli beaker has a recess that fits over the crystal in a gamma-counter, resulting in a much better counting geometry. A sensitivity of about 30 $pCi \cdot L^{-1}$ can be obtained for a 10-minute count.

For still better sensitivity, one must turn to liquid scintillation counting which has been described by Prichard and Gessel[31] and others. In this method, a 10 ml water sample is collected in a syringe and later injected into a 20 mL glass scintillation vial along with 5 mL of scintillation fluid. The vial is then counted in a liquid scintillation counter. A sensitivity of 10 $pCi \cdot L^{-1}$ is obtained in a 10-minute count. Because the syringes are small, many samples can be collected conveniently. Also, the use of an automatic counter simplifies analyses in the laboratory.

VENTILATION

The classic method for measuring ventilation rate in a building employs a tracer gas. Many gases have been used; we have used helium and radon itself, while others have used krypton-85, sulfur hexafluoride, hydrogen and ethane. One approach is to inject a slug of gas into a room, mix it thoroughly, and then monitor the reduction of concentration with time. Ideally, the reduction is exponential and the rate of air exchange can be obtained directly from the slope of the curve.

Another approach, which seems to be gaining in popularity, is to inject the gas at a slow, uniform rate until a steady-state concentration is attained. The ventilation rate in that case is calculated from the rate of gas input and the resultant air concentration.

The tracer gas technique has been verified by ventilation engineers in decades of use but its use in connection with radon studies must be cautious because the sources and distribution of radon are complex.

SUMMARY

Sufficiently varied techniques have been developed to meet most of the requirements for monitoring radon that are likely to be encountered. Unfortunately, the availability of commercial instrumentation is lagging behind the technological advances, often necessitating procurement from non-commercial sources, an inefficient and time-consuming procedure.

Since radon monitoring techniques differ in their capabilities, selection must be based on the needs of the particular mission. A basic decision is whether to measure radon or radon daughters. Other questions to be decided concern the duration of monitoring and whether time-variation of concentration or average concentration is needed.

Time variation and spatial variation, inherent characteristics of radon both indoors and in outside air, must be taken into account in the conduct of any monitoring program. In the measurements of average concentration, they are

much greater sources of error than instrument error which usually is small.

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RADON AND RADON DAUGHTERS IN BUILDINGS: A SURVEY OF PAST EXPERIENCE

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INTRODUCTION

The potential for elevated levels of radon and progeny in inhabited structures has been recognized for many years. This potential has been realized in several locations when materials bearing relatively high levels of radium have been incorporated into building or fill materials in, under and around structures. Noteworthy of these situations are Grand Junction, Colorado, the phosphate mining area of Central Florida, Elliot Lake, Ontario, and many instances of the incorporation of natural activity in building materials[1,2]. In addition, recent studies have called attention to elevated radon and progeny levels in structures due to the release of radon from water used in the structure[3-5].

There are considerable radon and progeny data available on individual structures in these and other areas. However, the amount on information available on the factors which combine to establish the measured radon and progeny levels is sparse at best. In particular, the things which are of most interest to this forum, i.e. data on deterministically relating structural characteristics to radon and progeny levels in buildings are largely nonexistent. This information is becoming increasingly important as health effects data suggest establishing exposure guidance at relatively low levels.

Since energy conservative designs or certain building materials may contribute to radon guidelines being exceeded, this clearly indicates the need for a predictive model including all pertinent parameters.

PAST EXPERIENCE

This presentation will focus mainly on the studies conducted in the Central Florida phosphate mining area. This is due to the limited time available for this presentation and because results of the previously mentioned studies have been available for quite some time.

Florida Phosphate Studies

In June 1975, the Environmental Protection Agency, in conjunction with the Florida Department of Health and Rehabilitative Services (DHRS) and the Polk County Health Department, initiated a pilot study to examine the radiological impact of structures built on reclaimed phosphate land. The results of these studies have been reported in various publications[6-8].

The phosphate ore in the Central Florida land-pebble phosphate area contains between 40 and 50 pCi of radium-226 per gram of ore. The ore is normally covered with a sand overburden approximately 3 to 15 m thick, containing from 1 to 2 pCi/g of radium-226.

The standard mining practice in the Florida land-pebble phosphate fields is to strip the overburden and mine the phosphate ore with drag-lines. The reclamation process consists of filling the strip-mined areas with overburden removed from adjacent areas and some byproducts of the ore beneficiation process. The land area thus reclaimed may contain from 1 to 50 pCi/g of radium-226 depending on reclamation techniques employed. The radium concentrations in reclaimed land have been found to vary widely both horizontally and vertically within relatively small areas.

The amount of reclaimed phosphate mining land in Florida has been estimated at approximately 25,000 acres (100 km^2). Approximately 30% of this land contains residential structures and 8% commercial structures. The remainder is used for parks, farming, and grazing. It has been estimated that an additional 70,000 acres (280 km^2) of reclaimed land may become available within the next 30 years. In addition, there may be a large area of land where phosphate deposits containing radium exist near the surface.

Two measurement techniques were employed in the Florida phosphate studies, namely, the Radon Progeny Integrating Sampling Units (RPISU) [9] and Track Etch* films[10].

The land on which the tested structures were constructed was classified according to four categories: non-mineralized (no deposits), mineralized (deposits present but unmined), reclaimed, and other (missing or incomplete information). Table 1 gives the results of the EPA-DHRS results for RPISU data for the 133 occupied structures[7]. (Over 200 structures were studied, but for various reasons only 133 were selected for analysis.)

Table 1

EPA and DHRS Radon Progeny Levels and Distribution by Land Category

Working Level (gross)	No. of Structures	%
> 0.05	8	6
0.03-0.05	12	9
0.01-0.03	26	20
< 0.01	87	65

Land Use	Mean WL	N	WL< 0.01	0.01<WL <0.03	0.03<WL <0.05	WL> 0.05
Reclaimed	0.017	93	61%	18%	13%	8%
Mineralized	0.015	9	56%	33%	11%	0%
Non-Mineralized	0.003	29	97%	3%	0%	0%
Unknown	0.009	2	0%	100%	0%	0%

A statistical analysis of these data indicate that working levels in the structures on non-mineralized land are different from those on reclaimed land at the 99% confidence level. Further, working levels in structures on mineralized land are not different from reclaimed land at the 90% confidence level.

The track etch data from the phosphate study have been analyzed by Robinson[11] to draw some tentative conclusions on the manner in which certain structural characteristics affect radon progeny levels. Since the study was not designed to provide the data for such a correlation, the tentative nature of this analysis must be emphasized. Table 2 summarizes the data used in this analysis. Because 90% of the structures studied were single-family residences only those structures are analyzed. The track etch data were regressed against external gamma exposure rates measured at each tested structure in order to account for the

Table 2

Summary of Relevant Characteristics for Single-Family Residences

			Track Etch Data	
	No.	%	Mean Tracks-hr mm^2	Standard Deviation Tracks-hr mm^2
Total	182	100	0.0025	0.0028
BUILDING STRUCTURE				
Basement or multi-level	11	6	0.0009	0.0011
Single-level slab	148	81	0.0029	0.0029
Single-level crawl space	15	8	0.0005	0.0004
Multi-level crawl space	3	2	0.002	0.0006
Trailer	3	2	0.001	0.0005
Unknown	2	1	0.004	0.0003
BUILDING MATERIALS				
Masonry	161	88	0.0028	0.0029
Non-Masonry	16	9	0.0009	0.0008
Trailer	3	2	0.0010	0.0005
Unknown	2	1	0.0004	0.0003
AIR CONDITIONING				
Yes	147	81	0.0029	0.0029
No	33	18	0.0008	0.0009
Unknown	2	1	0.0033	0.0011

* Service mark, General Electric Company

fact that structures with particular characteristics are built on lands with varying amounts of radium-226. The external gamma exposure should be an indication of soil radium-226. The regressions include a binary variable to represent the housing characteristic of interest.

Table 3 summarizes the results of these analyses.

Table 3

Effect of Working Levels of Indicated Housing Characteristics

	Significant?	
Crawl Space vs Slab-on-Grade	Yes	Lower WL for Crawl Space
Multi-Level vs Single-Level	Yes	Lower WL for Multi-Story
Air Conditioning vs Non-A/C	Yes	Lower WL for Non-A/C
Masonry vs Non-Masonry	Yes	Lower WL for Non-Masonry

Due to the limitations of study design, the small number of structures having some characteristics and the "looseness" of some of the regressions, these correlations must be tentative. However, these analyses do verify effects expected from physical principles.

Structures built with crawl spaces appear to have lower radon progeny levels than slab-on-grade ones. This would be expected due to the dilution of radon concentration prior to diffusion into the structure.

Multi-level structures apparently have lower working levels than single level ones. The radon input per unit time into a structure should be proportional to the product of the radon flux of the soil on which it is constructed and ground floor area of that structure. With adequate mixing, the radon (progeny) concentration is then proportional to the radon input and the volume of the structure, for a constant ventilation rate. Thus, a multi-level structure with a higher volume-to-foundation ratio would be expected to exhibit lower radon progeny levels than a single-level one.

The warm, humid climate in the area of this study makes it necessary to cool structures a large portion of the year. If the house is not air-conditioned it is probable that the cooling is provided by natural or forced ventilation. The fact that the working levels in air-conditioned structures appear to be higher than non-air-conditioned is an expected result due to the increased ventilation in non-air-conditioned houses.

There are many reports of increased radon and progeny levels in structures of masonry type materials. These reports tend to support this effect in the studied structures.

Environmental Measurements Laboratory Study - New Jersey - New York Area [3,12]

This study was performed by George and Breslin over a two-year period in 21 residences, mostly single-family one- or two-story wood-frame or brick construction with full basements (majority finished). These houses were built on land with radium-226 concentrations of approximately 1 pCi/g which is within the range of "background" (unenhanced) values. The houses were occupied during the study. The mean working level in the cellars of the studied structures range from approximately 0.002 to 0.026 WL. The first floor values range from 0.002 to 0.013 WL.

Simultaneous week-long measurements of radon and progeny were made in the cellars and on the other floors. These measurements were made periodically and distributed over the study duration. The average first floor to cellar radon concentration ratio of individual paired values was 0.53 $\pm$ 0.22. The average ratio of individual paired values of working levels for first floor to cellar was 0.56 $\pm$ 0.30. The average ratio of second to first floor working level was 1.06 $\pm$ 0.29.

These week-long measurements were highly variable, reemphasizing the need for long-term rather than "grab-sample" type monitoring in occupied structures.

A more recent publication for this study[3] shows that average radon concentrations in cellars are correlated with measured radon exhalation from cellar floors. It is further suggested that radon released from water used in the structure has a discernible effect on cellar radon concentrations.

Other Studies

Many other studies of measurements of radon and progeny in structures have been reported [13-

19]. Many of these studies have been summarized by Harley[2]. The source of radon exposure in most of these studies is from radium-226 in the construction materials. A wide range of materials has been implicated and resulting radon progeny levels above the anticipated guidance level have been reported. In most cases, these studies have one or more of the following limitations:

a) They were conducted under artificial ventilation conditions;
b) Measurements were of short-term or grab grab samples; and
c) They were directed only toward the effects of a given construction material with insufficient detail for general applicability.

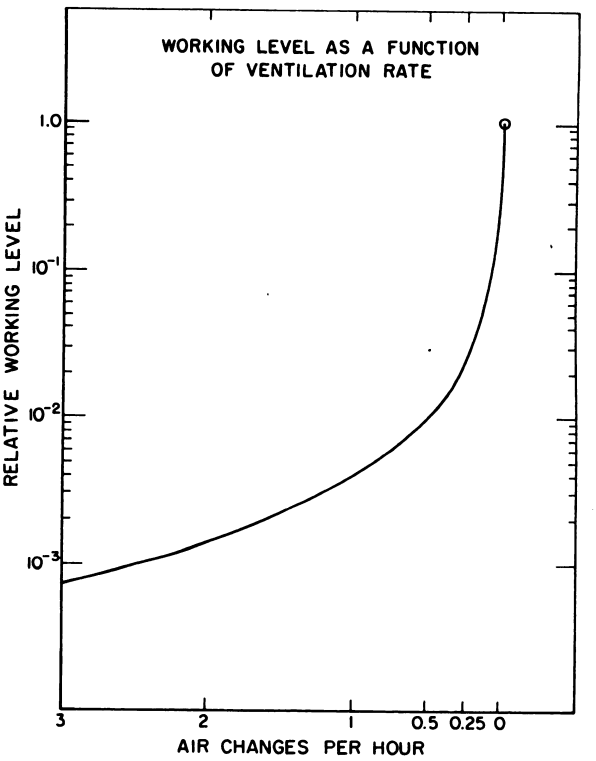
VENTILATION AND INFILTRATION

For a given radon input into a structure the radon and progeny concentrations in that structure are highly dependent upon the ventilation and infiltration (henceforth called ventilation) rates within that structure as shown in Figure 1. The magnitude of these rates is a function of wind speed, pressure difference between inside and outside, type of construction, workmanship, condition of the building and the activities of the occupants.

The most common method of determining ventilation rate is by releasing a tracer gas into the structure and measuring its concentration as a function of time. A literature survey of published data was performed by Handley and Barton [20]. They found the average annual ventilation rate of most occupied single-family houses falls in the range 0.5 to 1.5 air changes per hour.

Ventilation measurements made in a modern test house, constructed to minimize infiltration, indicate ventilation rates from 0.22 to 0.4 air changes per hour, depending on wind speed [21]. The test house was unoccupied; however, utility usage was simulated. Other unpublished measurements on similarly constructed "energy-conservative" houses indicate ventilation rates of from 0.1 to 0.2 air changes per hour.

Figure 1



Occupancy factors must not be ignored when making ventilation measurements or in using some published value for calculation purposes. For example, in a test house in Florida, the central air-conditioning fan was allowed to operate continuously for three hours. There was a subsequent reduction in working level by a factor of ten and a reduction in radon by a factor of five from that of a closed, unoccupied condition. This represented an effective ventilation rate change from 0.5 to 2.5 air changes per hour [8].

CONTROL TECHNOLOGY

A study funded by the Colorado Department of Health evaluated radon progeny control measures including dilution by ventilation, air cleaning with HEPA filters and air cleaning by electrostatic precipitation[22]. This report concluded that dilution by ventilation provides the most cost-effective method for radon progeny reduction, closely followed by elastrostatic precipitation.

Fitzgerald et al.[23] evaluated various radon progeny control measures applicable to the Florida phosphate situation, i.e. radon entry through structure floor.

The criteria for selection of an optimal control measure were: (1) operative passivity; (2) uniform effectiveness over the lifetime of a structure; (3) a one-time reasonable cost upon implementation; and (4) minimum impact on the lives of future occupants.

The evaluation is primarily, of necessity, based on theoretical predictions as very little experimental data exists which is directly applicable to the control of radon and progeny in structures. An exception to this lack of experimental efficacy verification is for the polymeric sealants [24, 25].

Table 4 summarizes the control measures considered in Ref. 23 with the authors' listing of advantages and disadvantages.

Table 4

CONTROL MEASURES: ADVANTAGES AND DISADVANTAGES

CONTROL MEASURE	ADVANTAGES	DISADVANTAGES
FULL MEASURES		
VENTILATED CRAWL SPACE	1-PASSIVE, LIFE-OF-HOUSE MEASURE 2-HIGH EFFECTIVENESS WITH PROPER VENTILATION	1-EXPENSE ASSOCIATED W/CHANGES IN HOME'S STRUCTURAL SPECIFICATIONS 2-BUYER'S OBJECTIONS ON AESTHETIC OR OTHER GROUNDS 3-HIGHER ELECTRICAL COST FROM INFILTRATION
POLYMERIC SEALANT	1-PASSIVE MEASURE 2-WITH PROPER APPLICATION, HIGH EFFECTIVENESS ACHIEVED AT RELATIVELY LOW COSTS 3-SEALANT PROVIDES ADDITIONAL WATER-PROOFING	1-UNCERTAINTY AS TO LONGEVITY OF SEALANT EFFECTIVENESS 2-THOROUGH APPLICATION NECESSARY FOR EFFICIENT RADON REDUCTION
EXCAVATION AND FILL (W/NOMINAL FILL COST)	1-PASSIVE, LIFE-OF-HOUSE MEASURE 2-NO ALTERATION IN HOUSE PLANS OR CONSTRUCTION PROCEDURES NECESSARY	1-EFFECTIVENESS BASED ON THEORY ALONE 2-PROBLEMS ASSOCIATED WITH IMPROPER COMPACTION OF FILL 3-RADON INFILTRATION ALONG PIPING LAID UNDER SLAB
EXCAVATION AND FILL (AT COMMERCIAL RATES)	SAME AS ABOVE	SAME AS ABOVE EXCEPT FOR HIGH COST ASSOCIATED WITH FILL
ELECTRONIC & AIR EXCHANGER	1-SYSTEM DURABLE 2-LITTLE MAINTENANCE REQUIRED 3-PARTICULATE CONCENTRATION REDUCED IN HOME	1-NON-PASSIVE 2-HIGH COST INVOLVED WITH OPERATION 3-OZONE RELEASED BY PRECIPITATOR
IMPROVED SLAB CONSTRUCTION (8" SLAB)	1-PASSIVE, LIFE-OF-HOUSE MEASURE 2-VERY LITTLE ALTERATION IN CONSTRUCTION PLANS REQUIRED	1-CRACKING IF IT OCCURS, WOULD NEGATE MUCH OF EFFECTIVENESS
LIMITED MEASURES		
ELECTRONIC AIR CLEANER	1-MINIMAL ELECTRICAL COST AND MAINTENANCE NECESSARY 2-PARTICULATE CONCENTRATION REDUCED IN HOME	1-NON-PASSIVE 2-OZONE RELEASED BY PRECIPITATOR
HEPA FILTERS	1-EFFECTIVE AT SMALL PARTICLE SIZE RANGE 2-NON MECHANICAL PARTS, NO MAINTENANCE NECESSARY	1-NON-PASSIVE 2-HIGH COST OF FILTERS 3-HIGH PRESSURE DROP (AS AIR FLOW RATE INCREASES, EFFICIENCY DECREASES)

Table 5 gives the authors' estimated efficacy and costs for the considered techniques. The authors conclude that the inclusion of crawlspace in the construction of a residence is the most cost-effective technique which satisfies the criteria for optimal radon progeny control.

This study by Fitzgerald et al. also calls attention to the potential for increasing lung doses due to increased free ion fractions with techniques employing high air exchange rates or selective removal of aerosols through filtration.

When using control techniques that decrease the diffusion of radon through the floor, the potential increase in whole-body gamma exposure due to radon daughter build-up and decay in and under the floor must be considered[26].

CONCLUSION

This paper has attempted to review the literature on the factors that affect the radon and progeny levels in structures. Although a seemingly large volume of data is available on this subject, it falls far short of being sufficient, in this author's opinion. We know only in a qualitative sense the deterministic factors affecting these levels. We are not able to answer the principal question: "What will be the radon progeny concentration in a structure constructed of given materials, in a given manner on a known location?" Hopefully, this meeting is the needed impetus to begin to address this question.

Table 5

AVERAGE COST PER PERCENT REDUCTION OF RADON DAUGHTER LEVELS

CONTROL MEASURE	ESTIMATED PERCENT RADON PROGENY REDUCTION	TOTAL COST (30–50 YRS)*	AVERAGE COST/PERCENT REDUCTION	RANK FULL MEASURE**	RANK LIMITED MEASURES
AIR CLEANERS:					
HEPA	40%	$1725–1925	$43–48($45)	–	5
ELECTRONIC	40%	800– 900	20–23($21)	–	2
ELECTRONIC & AIR EXCHANGE	60%	3000–3325	50–55($52)	5	6
VENTILATED CRAWL SPACE	80%+	450	6($ 6)	1	1
POLYMERIC SEALANT	80%	400–1300	5– 16($ 10)	2	3
EXCAVATION AND FILL (TO 10' DEPTH):					
COMMERCIAL FILL RATE	80%	2100–3700	26– 46($ 36)	4	–
W/NOMINAL FILL COST	80%	1595–1850	20– 23($ 22)	3	–
(TO 5' DEPTH):					
COMMERCIAL FILL RATE	40%	1100–1900	28– 48($ 38)	–	4
W/NOMINAL FILL COST	40%	800– 950	20– 24($ 22)	–	2
IMPROVED SLAB CONSTRUCTION	80%+	450	6($ 6)	1	1

*WITH 6% DISCOUNT RATE.
**MEASURES WITH ESTIMATED REDUCTION OF EFFICIENCIES OF 50% OR MORE.

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RESIDENTIAL BUILDING TECHNOLOGY TRENDS AND INDOOR RADON AND RADON DAUGHTER CONCENTRATIONS

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ABSTRACT

Radon-222, a radioactive gas, is a decay product of naturally occurring Ra-226. Small amounts of Rn-222 and its further decay products are present in the indoor and outdoor environments. Since the energy crisis of 1973-74, residential building trends have been in the direction of providing more energy-conserving residences. In addition, higher energy costs and energy supply uncertainties have motivated occupants to operate residences in more energy-conserving ways. This paper surveys several of these trends as they relate to the resulting concentrations of Rn-222 and its daughters in the residential environment. It is concluded that energy-conserving measures tend to increase indoor potential alpha energy levels associated with radon progeny up to 10 times those typical of pre-energy crisis construction and operational strategies. These higher levels may indicate a future additional human health hazard.

Key words: Construction materials; earth berms; energy conservation; indoor air quality; natural gas; passive solar energy; radioactivity; radon; ventilation

INTRODUCTION

Radium-226 is widely distributed in minerals over the earth. It has a half-life of 1600 years so, for practical purposes, its activity is invariant with time. Ra-226 decays to gaseous radon-222, which can diffuse through many materials and become airborne. Rn-222, in turn, decays to give a chain of solid decay products, with short half-lives until Pb-210, which has a 22-year half-life. Table 1 lists these decay products.[1]

Table 1. Relevant Ra-226 Decay Products[1]

Radionuclide		Half-life	Potential alpha energy per atom (MeV)
Radon	Rn-222 $\downarrow\alpha$	3.8 d	-
Polonium	Po-218 $\downarrow\alpha$	3.1 min	13.68
Lead	Pb-214 $\downarrow\beta$	27 min	7.68
Bismuth	Bi-214 $\downarrow\beta$	20 min	7.68
Polonium	Po-214 $\downarrow\alpha$	<1 s	7.68
Lead	Pb-210	22 y	-

Thus, if Rn-222 diffuses into indoor air, these radioactive progeny may remain airborne by becoming attached to dust particles, which could be deposited in the lungs of exposed persons. Alpha particles emitted by attached or unattached (to aerosol) radon progeny could be health hazards depending on their concentrations in the body.

The health effects of radon daughters depend on the form in which they occur since the form affects their penetration into the lungs. They are found in two distinct fractions[2]: an unattached fraction composed of free ions and atoms, and an attached fraction of daughters associated with aerosols. Most of the respired unattached fraction (60-70%)[3] is removed by nasal deposition but part can penetrate as far as the bronchial region. Because of the high diffusion coefficient, any remaining unattached daughters deposit on the bronchi, and there is little deeper penetration into the lung. The unattached fraction may be a cause of lung

cancer in uranium miners, which presumably originates in the underlying basal cells of the bronchi.[4]

The attached fraction is found predominantly on respirable particulates (activity median diameter between 0.1 and 0.2 μm).[2] Very little of this fraction is removed by the nose, and the particles can penetrate into the pulmonary region of the lung.[5,6]

A measure of potential alpha energy is the working level (WL), defined is any combination of short-lived radon daughters per liter of air yielding 1.3×10^5 MeV of alpha energy upon decay to Pb-210. If Rn-222 and its daughters were in equilibrium, this would correspond to 100 pCi/L (3700 Bq/m^3) of Rn-222. The equilibrium factor (F) is used to convert between Rn-222 concentration and potential energy. It is defined as the ratio of actual potential alpha energy to that if daughters were in equilibrium with Rn-222.

$$F = (100\ pCi \cdot L^{-1}/WL \text{ or } 3700\ Bq/m^3) \times \frac{\text{potential alpha energy}}{\text{Rn-222 concentration}}$$

Atmospheric diffusion and wind dilute Rn-222 and progeny activities in outdoor air. Rn-222 in indoor air is diluted by air leakage (infiltration) and ventilation. Fallout of dust particles with attached daughter products may also decrease airborne activity.

There are four identifiable sources of Rn-222 gas which can be introduced into indoor air and would increase the indoor activity with respect to that outdoors:[1]

1. Soil-produced Rn-222 in the ground in contact with a residence. Rn-222 can diffuse through the earth, basement walls and floors at a rate determined by soil Ra-226, U-238 (an "ancestor" of Ra-226) and moisture content, porosity, and temperature.
2. Construction materials. Concrete, brick, stone, plaster, sand and gravel contain Ra-226 in varying concentrations. Wood products do not contain sufficient amounts of Ra-226.
3. Residential water supplies from ground water. Such water contains dissolved Rn-222 which can outgas in the residence as the water is used. Bathing, and washing of clothes and dishes release a significant fraction of the Rn-222 into the air.
4. Natural gas. Natural gas contains varying concentrations of Rn-222. Gas cooking and unvented space heating are the significant sources. Vented gas appliances would presumably also vent most of the Rn-222 and daughters from this source from the residence.

Commonly accepted building construction in the past has resulted in infiltration rates in residences sufficiently high that no indoor health hazards have been identified, with the exception of a few cases where houses were built unusually radioactive material obtained from uranium mill tailings[7] or phosphate ores.

POST-ENERGY CRISIS TRENDS

Tighter Construction

Construction practices prior to 1973-74 resulted in uncontrolled air leakage (infiltration) of about 0.5 to 1.5 air changes per hour (ach).[10] Air exchange rates increase with increasing wind velocity and increasing indoor-outdoor temperature difference for any building. Air exchange rates below 0.5 ach can occur even in buildings with loosely constructed envelopes if there is little indoor-outdoor temperature difference and low wind speed, but less frequently than in tighter buildings. Construction details and workmanship are responsible for significant variations among residences.

Infiltration may account for approximately 25% of the heating and cooling cost of a residence[11] but its share would increase if insulation levels were increased without a corresponding reduction in air leakage. Thus application of current technology also results in lower infiltration rates, and this trend will undoubtedly continue. One practical technique to reduce infiltration is to wrap the framing of the residential exterior walls and ceiling with a polyethylene film and apply interior dry wall or other surfacing over it. Another trend is toward more effective weatherstripping of doors and windows to reduce infiltration. Homes have been built which have measured infiltration rates as low as 0.1 ach, and perhaps 0.5 ach will be a

future accepted norm partly because water condensation may be a problem at lower air exchange rates.

If air exchange rates in residences were reduced from about 1 to 0.25-0.5 ach, Rn-222 activities could increase 2- to 4-fold. Decreased air exchange rates also affect total potential alpha energy, the equilibrium factor, the attached fraction/unattached fraction ratio, and the rate of particulate removal by surfaces: walls, floors and furniture.[12-14] Predicting the extent of the changes in these parameters is not simple.

Fig. 1 shows the effect of changing ventilation on total potential alpha activity in a residence, as calculated by a computer model developed at the Center for Building Technology of the National Bureau of Standards.[15] The model

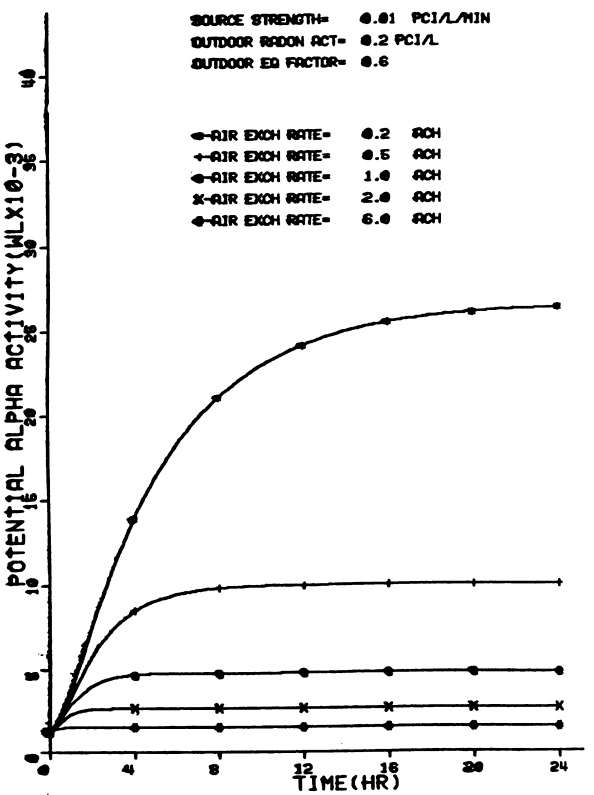

Figure 1. Time course of potential alpha activity at several air exchange rates.

assumes typical indoor Rn-222 emanation rates, typical outdoor Rn-222 and daughter levels,[1] and includes the increase in equilibrium factor indoors due to lowered ventilation rates, but does not include particulate settling. (The equilibrium factor increases with decreased ventilation because more of the Rn-222 has a chance to decay before being exhausted from the building). Since most of the potential alpha activity is associated with the attached fraction, and this fraction closely parallels total activity, the figure shows very well the increased dose to the pulmonary region in the absence of any air cleaning.[1]

When the air exchange rate is reduced from 1 to 0.5 ach, steady-state potential alpha activity increases 2.1-fold. A reduction to 0.2 ach causes a further 2.1-fold increase (the 8-hour, not the steady-state, value is used for 0.2 ach because it is doubtful that the air exchange rate would remain constant any longer). Potential alpha activity increases from 0.0048 WL at 1 ach to 0.010 WL at 0.5 ach, and to 0.021 WL at 0.2 ach. The Surgeon General of the U. S. Public Health Service recommended remedial action at Grand Junction, Colorado, at levels between 0.01 and 0.05 WL above background.[1]

Although total daughter activities rise markedly when air exchange rates are lowered, increased aerosol levels and residence times of air in buildings also could greatly reduce the unattached fraction[14] so it is difficult to predict whether the unattached progeny concentration would increase or decrease.

Homeowners might be encouraged to replace combustion-fired with electric appliances, or to install air-cleaning equipment like electrostatic precipitators or other high-efficiency filters. These measures might reduce total potential alpha activity in the unattached fraction. Table 2 summarizes what is known about the effects of decreased ventilation on radon levels in indoor air.

Table 2

Effects of Air Cleaning on Radioactivity due to Radon-222 and Progeny

	Air Cleaning	No Air Cleaning
Total potential α energy	unknown	increases
Equilibrium factor	unknown	increases
Unattached fraction	increases	decreases
Total potential α energy on unattached fraction (and effect on tracheo-bronchial region)	unknown	unknown
Attached fraction	decreases	increases
Total potential α energy on attached fraction (and effect on pulmonary region)	unknown	unknown

Increased Use of Thermal Storage

It is expected that much more use will be made of thermal storage in homes to save energy. Solar-heating systems and possible off-peak power-pricing strategies could encourage use of thermal storage. Massive concrete, brick or stone walls, floors, and room dividers are likely to find increased use in residences. These may significantly increase Rn-222 activity inside homes.

Since building materials often emit less Rn-222 than soil,[1] the main effect of increased use of massive concrete, brick and stone walls on indoor Rn-222 levels may be through the tighter envelopes that result. Figure 2 shows the results on potential alpha energy levels when the source strength is doubled from 0.01 to 0.02 pCi/L·min (6×10^{-3} to 0.012 Bq/m^3·sec) and and the air exchange rate is halved from 1 to 0.5 ach.

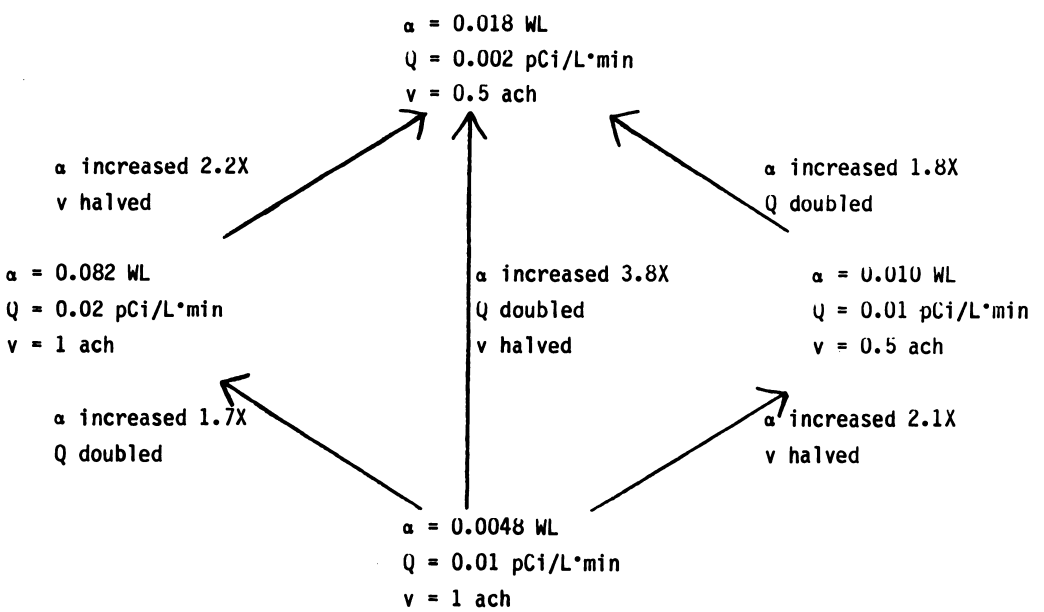

Figure 2. Increase in potential alpha energy (α, WL) with increasing source strength (Q, pCi/L·min) and decreasing air exhcange rate (v, ach).

The effects of changes in building materials are illustrated by Swedish experience.[17] Homes built before 1930 had average Rn-222 activity levels of about 0.54 pCi/L (20 Bq/m^3) compared to 0.86 pCi/L (32 Bq/m^3) for those built between 1941 and 1950 and 2.9 pCi/L (107 Bq/m^3) for those built between 1971 and 1975. Most of the 3.3-fold increase between 1941-50 and 1971-75 can be attributed to the reduction in average air exchange rates from 0.8-0.9 to 0.3-0.45 ach, but some of it is due to changes in building materials. The contribution of building materials is seen more clearly in the 1.5-fold increase in Rn-222 activity between houses built before 1930 and those built in the period 1941-50. There was little difference in air exchange rates between these two groups of houses.

Earth berms

Another strategy which can save energy in buildings is partial burying to take advantage of the earth's insulating value and thermal storage. Many experimental buildings have been built where entire walls and even roofs are banked with earth. Partially buried walls would presumably be more common. In any case, increased earth in proximity to the building shell could increase Ra-226 activity near the shell, and larger quantities of Rn-222 could diffuse inward.

Economic Considerations

Higher energy costs as well as general inflation may cause a future reduction in residential size. If this occurs, the ratio of surface area to volume of the residence will increase, causing an increase in indoor Rn-222 activity arising from subsoil or building material. The increase in Rn-222 activity from decreased floor area and volume is calculated in Appendix A. Rn-222 emanation rates from building materials per unit volume increase about 11% for the extreme case of a 1-story house with no attic or basement where walls and floors contribute equal activities of Rn-222 per unit time and area, and the house volume (or floor surface) is decreased by 1/3 from 150 to 100 m^2.

In areas where the main Rn-222 source is natural gas or water, the effect of decreasing house size would be greater. If the same quantities of these materials were used inside the two houses above, there would be a 50% increase in Rn-222 activity from these sources. If the equilibrium factor were also raised, then the increase in potential alpha activity would be still greater. Occupants of residences could also be expected to alter their behavior to save energy and money, for example by reducing operation of the usually present ventilating fans in bathrooms and kitchens, or by not opening windows. These actions would reduce venting of Rn-222 and its daughters from water and natural gas sources and therefore increase indoor radioactivity levels.

POSSIBLE CONTROL STRATEGIES

If Rn-222 activity levels are known to be hazardous, there are several possible controls, but with economic penalties. For example, additional natural or mechanical ventilation air could be provided at the expense of additional energy use. Heat exchangers now being introduced in Europe, which transfer heat from the exhaust to incoming ventilation air, could also be employed at a capital cost penalty. High-efficiency air cleaners used on recirculated air could reduce levels of particulates and attached radioactivity. These air cleaners are currently available, but are rather costly for widespread residential use. Surface coatings impervious to Rn-222 for basement walls and floors are being developed and tested.[18] Other architectural strategies might also be developed.

CONCLUSION

Several trends in housing construction and occupant strategy are likely to become permanent changes due to to increased costs and uncertain availability of energy. These trends suggest that higher activity levels of Rn-222 and its progeny will occur in future residences than in those built with pre-energy crisis practice. It is calculated that, under the most extreme circumstances, future potential alpha activities up to 10 times past levels may occur. Therefore, this problem should be investigated to obtain more quantitative data to further assess health hazards.

APPENDIX A

RN-222 EMMISSION AND REDUCTION OF HOUSE VOLUME OR FLOOR SURFACE

Let A_i = floor area (m^2) of house i. A_i is proportional to volume because height is assumed constant.

E_i = total Rn-222 emanation rate from exterior walls and floors per unit volume (pCi/L·min)

S_f = Rn-222 emanation rate per unit floor surface (pCi/min·m^2)

S_w = Rn-222 emanation rate per unit wall surface (pCi/min·m^2)

w_i = width (m) of house i

l_i = length (m) of house i

h = height (m) of house = 2.5 m

$\gamma = l_i/w_i$ = length/width ratio

$\delta = w_2/w_1 = l_2/l_1$ = ratio of linear dimensions of house 2 to house 1.

A_f = floor surface area (m^2)

A_w = exterior wall surface area (m^2)

V = volume (m^3)

h, γ and δ are assumed constant when floor surface is reduced, i.e. the floors of both houses are assumed to retain the same geometry. The ceiling and interior wall partitions are assumed to contribute a negligible amount of Rn-222 activity since they probably contain little concrete.

$$E_i = \frac{A_f S_f + A_w S_w}{V}$$

$$= \frac{w_i l_i S_f + 2h(w_i + l_i) S_w}{w_i l_i h}$$

$$= \frac{1}{h} S_f + 2\left(\frac{1}{l_i} + \frac{1}{w_i}\right) S_w$$

$$= \frac{1}{h} S_f + 2\left(\frac{1}{\gamma} + 1\right) \frac{1}{w_i} S_w$$

Therefore:

$$\frac{E_2}{E_1} = \frac{1 + 2\left(\frac{1}{\gamma} + 1\right) \frac{h}{\delta w_1} \frac{S_w}{S_f}}{1 + 2\left(\frac{1}{\gamma} + 1\right) \frac{h}{w_1} \frac{S_w}{S_f}}$$

Let $\gamma = 4/5$, corresponding to a reduction in V of about 1/3. Assume $A_1 = 150\ m^3$, then $A_2 \approx 100\ m^3$. Usually, $S_w < S_f$.[1] If S_w is very small, then $E_2/E_1 \approx 1$. Take $S_w/S_f = 1$ as an extreme case. Three cases are $\gamma = 1$ (square house), $\gamma = 1.5$ (length = 1.5 x width), and $\gamma = 2$ (length = 2 x width). These yield increased radon concentrations due to the structure itself of 11.2, 11.3 and 11.6%, respectively. A sample calculation is given for the first case:

$A_1 = w^2$

$w_1 = 12.247$ m

$h/w_1 = 0.204$

$E_2/E_1 = 1.112$

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EPA APPROACH TO ESTIMATING HEALTH RISKS OF RADON/RADON DAUGHTER EXPOSURE

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The EPA has used the results of epidemiological studies of uranium and other underground miners to obtain risk coefficients for estimating the health impact (fatal lung cancer) due to radon/radon daughter exposure. Animal exposure data, although now showing a dose response curve, is of little help due to differences in human-animal lung morphometrics, dosimetry and the cells of origin of lung cancer. It is not always appreciated that animal lung cancers differ in histological classification from common human lung cancers. U.S. uranium miner data is likewise of little use since exposures generally greatly exceeded several hundred cumulative working level months (CWLM).

EPA has derived an average relative risk coefficient of 3% increase in lung cancer per CWLM exposure from an evaluation of Ontario, Czech, and Swedish miner data. These data, aggregated for all ages of miners, show for exposures less than a few hundred CWLM a linear dose response curve. Apparent increases in fatal lung cancer range from 1% per CWLM at high CWLM to 4% or more per CWLM at low CWLM exposures.

Since published data show no striking differences between aerosol characteristics in mines, outdoors, or inside, the major difference in exposure between miners and a general population is the volume of air respired. A miner engaged in a mixture of light and heavy work inhales about 3×10^5 liters of air a month, a reference adult in the population about 2.2 times greater volume. In a 1 WL atmosphere, a miner would be exposed to 12 CWLM per year, a reference adult about 27 CWLM. However, Americans spend only about 75% of their time in their residence; so a residential exposure to 1 WL indoor radon/radon daughter concentration corresponds to 20 CWLM per year.

Using a 3% CWLM relative risk model and residential exposure to 0.02 WL, the following population risk estimates were made in an EPA report.[1]

Lifetime Risk of Lung Cancer per 100,000 Persons
Due to Lifetime Residency in Structures with
an Average Radon Daughter Concentration of 0.02 WL

Relative Risk	Excess Cancer Deaths	Total Years of Life Lost
Adult and Child Sensitivity Equal	2,000	30,000
Child Sensitivity 3 times Adult	3,000	50,000
Absolute Risk (10 deaths/CWLM for 10^6 person-years at risk)	1,000	27,000

In perspective, the expected lifetime lung cancer mortality in the U.S. population is 2,900 per 100,000 persons in a population having the age specific mortality pattern of the 1970 U.S. population.

A detailed analysis of the rationale for estimating radon/radon daughter health effects is given in the EPA publication. [1]

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THE ENVIRONMENTAL WORKING LEVEL MONITOR (EWLM)

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The Environmental Working Level Monitor (EWLM), designed and developed at Argonne National Laboratory for the EPA, is a fully automated, microprocessor- controlled, portable instrument, which allows monitoring of airborne radon-daughter concentrations and the Working Level (WL). It has a monitoring range of 10^{-3} to 10 WL or a lower limit of approximately 0.1 pCi per liter for the radon-daughter concentrations. Thus the instrument is capable of measuring the normal background levels of these nuclides. The EWLM requires 9 minutes to make and record a measurement. It can be programmed to automatically take and record 192 measurements at a minimum time interval of one measurement every 10 minutes. The time interval between measurements can be programmed for any period of time greater than 10 minutes, such as one measurement every hour for 8 days. The starting time and completion time are also programmable.

To provide these measurements, the EWLM uses a diffused junction detector for the alpha particles and a NE102 scintillator optically coupled with a 10-stage photomultiplier for the beta particles.

A single channel analyzer resolves the 6.0 MeV RaA (^{218}Po) from the 7.69 MeV RaC' (^{214}Po) alpha particles. The separated RaA and RaC' (^{218}Po and ^{214}Po) counts are stored in the computer memory along with the total (beta and gamma) counts from RaB and RaC (^{214}Pb and ^{214}Bi). These three stored counts are used by the internal computer to calculate the radon-daughter concentrations and the WL. The EWLM method does not make any assumptions about the equilibrium conditions between the radon daughters, but does assume, like all other methods, that the airborne radon-daughter concentrations are constant during the sampling period of three minutes.

The calibration of the instrument is quite complex. Briefly, it requires analysis of the same air sample by the EWLM method and by other methods which determine the radon-daughter concentrations and the WL from alpha counts observed during three different counting intervals.

The flexibility of this instrument with its computer-programmable options makes it a useful tool to study the effects of ventilation on the radon-daughter concentrations in buildings.

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TRACK ETCH TECHNIQUES FOR RADON MEASUREMENTS

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The Terradex Corporation has supplied several hundred thousand Track Etch® cups to the mining industry to measure radon levels in soil gas as a tool in uranium exploration. The Track Etch technique involves a simple dielectric detector which responds to alpha particle radiation damage tracks from radon and its daughters. The tracks are subsequently etched and counted to give a measure of the integrated alpha dose to the detector. Detectors can measure the integrated dose over exposure periods ranging from about one day to one year.

The Track Etch technique, originally invented for environmental measurements, has recently been significantly improved. New materials allow increased sensitivity, automatic readout and high resistance to environmental factors such as temperature and humidity. Membrane filters permit the exclusion of thoron gas from the cup and also of particulate radon daughters in the ambient atmosphere. Calibration of the Track Etch response to the standard radon atmospheres at the Environmental Measurements Laboratory in New York, have shown good reproducibility and will allow the Track Etch response to be reported directly in picocuries per liter.

The improved Track Etch system has the sensitivity and simplicity for inexpensive large scale screening of structures for radon levels.

* *

EPA RADIATION PROTECTION GUIDANCE FOR PHOSPHATE LANDS

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Ed. note: The speaker did not submit a summary for inclusion in this report. The EPA letter to the Governor of Florida setting forth the findings of its investigation of indoor radiation exposure due to radium-226 in Florida phosphate lands, and the recommendations for radiation protection of persons residing on phosphate lands has been published in the Federal Register, Vol. 44, 38664-70 (July 2, 1979). For further information, contact the speaker directly.

* *

RADON IN BUILDINGS: UNIVERSITY OF FLORIDA INTEREST AND ACTIVITIES

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Efforts are directed primarily to indoor radon associated with phosphate mining, land reclamation and by-product use. Emphasis areas include:

1. Developing a general overview of the situation;
2. Determining the relationship of indoor radon and radon progeny levels to land and materials history;
3. Development of methods for predicting future indoor levels for currently undeveloped lands; and
4. Proposing and verifying mining and land reclamation methods that minimize future radiological impact.

From the standpoint of developing criteria and remedial measures, three distinct situations exist:

1. Radioactivity in existing structures,
2. Future construction over existing mined but undeveloped land, and
3. Mining, reclamation and future development of currently unmined land.

Lands can be classified according to history - undisturbed, tailings reclaimed, low activity overburden, high activity overburden, etc. Each has its characteristic distribution of soil radium concentrations, and structure-land type categories have characteristic distributions of indoor radon progeny concentrations.

By estimating the number of structures in various land-structure type categories and summing distributions across categories, the distribution of indoor radon progeny concentrations was estimated for Polk County in the Central Florida phosphate mining region.

Predicting future indoor levels expected for currently undeveloped land is complicated by the many variables in this relationship. Soil surface radon flux, soil radium distribution and gamma radiation levels are being examined for sensitivity and usefulness as predictors.

For the case of currently unmined lands, the opportunity exists for limiting future indoor radon levels by measures applied during mining and reclamation. Mining companies are being encouraged to examine the feasibility of mining and reclaiming in a fashion that simulates the natural soil radium distribution - i.e. selectively placing the higher activity spoils and wastes under a cover of lower activity overburden. Future work will include field testing of the effectiveness of such procedures.

* *

PROPOSED PROJECT TO STUDY REDUCTION OF INDOOR RADON -222 CONCENTRATIONS THROUGH IMPROVED CONSTRUCTION TECHNIQUES

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Large areas of Central Florida, located primarily in Polk and Hillsboro Counties have been mined for phosphate bearing ore. Associated with this ore are significantly elevated concentrations of uranium and its decay products (about 30 to 60 times greater than those found in average soil and rock). As this ore is recovered large areas of waste land containing water filled trenches (phosphate pits), overburden, tailings and slimes from processing the ore are left behind. Much of the uranium and its decay products either remain in this waste land or are returned to it with the process wastes after the phosphate has been removed. A significant amount of this waste land has been and will be converted into beneficial and productive land for use in agricultural production, recreational and housing needs. Much of this land borders relatively large populated areas and is needed for home construction. In fact many homes have already be built on this reclaimed phosphate land and many are being built. There has been a great deal of data compiled by several government and private organizations concerning the problems associated with this construction. The major one being risk of increased cancer rates due to exposure to radon-222 daughter products. In the latest Environmental Protection Agency publication concerning the subject entitled Indoor Radiation Exposure Due To Radium -226 In Florida Phosphate Lands, the subject of control measures is discussed, and the following five types of control methods to reduce radon in homes are analyzed:

(1) Air particulate removal;
(2) Polymeric sealants;
(3) Ventilated crawl spaces;
(4) Excavation and fill methods; and
(5) Improved slab construction.

The five methods evaluated indicate an average probable reduction of radon decay product air concentration by about 80% but as of this date there have been no test structures built or tests performed to confirm any reduction figures.

The Polk County Health Department, with the cooperation of a local builder-contractor, has begun a project that should supply data on a number of control techniques to reduce radon concentrations in homes built on reclaimed phosphate lands. Several test homes will be built on a proposed tract of reclaimed phosphate land. The models will be of the following types:

(1) Improved slab construction - monolithic slab;.
(2) Monolithic slab with ventilation pipes to allow ventilation from under the slab to the exterior;
(3) Ventilated crawl space with wooden flooring at least twelve inches above ground surface; and
(4) Ventilated crawl space with prestress concrete floor at least twelve inches above ground surface.

Measurements taken at structure sites and in structures will include:

(1) Radium-226 analysis in the soil before construction;
(2) Radon exhalation rates from soil before construction;
(3) Soil density measurements before construction;
(4) Soil moisture measurements in conjunction with Radon exhalation rates;
(5) Radon exhalation rates after construction (near structures and underneath structures with crawl spaces);
(6) Radon concentrations in ventilated crawl spaces;
(7) Equilibrium measurements in crawl spaces;
(8) Radon concentrations in structures;
(9) Equilibrium measurements in structures;
(10) Working level concentrations in structures; and
(11) Gamma survey done over entire tract of land.

Upon completion of this project we hope to have identified at least one or two construction types that will reduce radon daughter concentrations to a level low enough to allow contractors and home builders to utilize the reclaimed phosphate land for further residential use.

* *

NRC INTEREST IN RADON IN BUILDINGS

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NRC interest in the issue of radon in buildings may be divided into three categories:

1. In the event that NRC is given the responsibility for regulating NARM (naturally occurring and accelerator-produced radioactive materials), the NRC staff may need to issue regulatory guidance to enhance human safety related to radon in buildings;
2. NRC licensing of consumer products used in buildings may need to take account of the elevated background population exposure due to radon in buildings and;
3. Models used to analyze radon in buildings may have application to analyses performed by NRC (for consumer products, transportation incidents, reactor accidents) and vice versa.

In July 1977, NRC published a task force report, "Regulation of Naturally Occurring and Accelerator-Produced Radioactive Materials," NUREG-0301. The task force recommended that the NRC seek legislative authority to regulate NARM, since these materials present significant radiation exposure potential and present controls are fragmentary and non-uniform at both the State and Federal level.

In the process of licensing consumer products such as ionization-type smoke detectors, tritium backlighted watches, and spark eliminators, the elevated radiation exposure levels in enclosed, interior spaces may need to be taken into account to avoid unnecessarily excessive cumulative exposures.

The NRC staff often needs to estimate the dose received by people in buildings resulting from sources of radioactive gases or aerosols--the source of which is either internal to the building (such as consumer products, defective products, or storage facilities) or external to the buildings (such as transportation or reactor incidents). One model used for transporation accidents[1] relates dose to an individual inside the building to the dose to an individual outside using parameters such as deposition, filtration, radioactive decay, air-recirculation, and infiltration factors. Another model[2] used for hazardous material spills relates the time history of concentration inside a building to the time history of concentration outside in terms of building volume and air change rate.

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[1] A. R. Ducharme, et. al., "Transport of Radionuclides in Urban Environs: a Working Draft Assessment", SAND 77-1927, May 1978, p. A-203.

[2] A. H. Rausch, et. al., "Continuing Development of the Vulnerability Model", CG-D-53-77, February 1977, Chapter 8.

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REPORT ON INTERESTS AT BROOKHAVEN NATIONAL LABORATORY

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The Biomedical and Environmental Assessment Division (BEAD) is part of the Brookhaven National Laboratory in Upton, New York. Most of the support of the division comes from the Office of the Assistant Secretary for the Environment of the Department of Energy. The division's mission is to help evaluate the human health and ecological impacts of alternative energy supply and use strategies.

The BEAD's interest in radon arises from two separate energy strategies only one of which is directly related to the subject of this discussion.

(1) Some building energy conservation methods will increase indoor levels of radon and its daughters. A former staff member of BEAD published work relating to the general problem of increased levels of indoor pollution before joining the National Bureau of Standards. No work has since been done at BEAD, nor is any contempleted, to pursue the specific topic of indoor radon health effects. Work does continue on general problems related to building energy conservation, indoor air pollution, and methods of monitoring exposure to indoor air pollution.

(2) Uranium mining, active mills, and mill tailings produce radon gas which raises outdoor levels. A paper is in preparation reviewing current knowledge about and estimates of the health effects from this source.

HEALTH TRADE-OFFS INVOLVED IN TIGHTENING HOUSES: INCREASED RADON EXPOSURE VS. DECREASED EXTERNAL AIR POLLUTION

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Ed. note: The speaker withdrew the summary submitted for inclusion in this report. For further information, contact the author directly.

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INDOOR RADON STUDIES AT LAWRENCE BERKELEY LABORATORY

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Space heating and air conditioning account for roughly 20% of the primary energy used in the United States. Rising costs and supply uncertainties are motivating builders and owners to reduce building energy consumption. Substantial energy savings can be achieved by reducing the rate at which outside air enters a building. This can lead to increased exposures to radon daughters and other indoor generated contaminants. The Building Ventilation and Indoor Air Quality Group at Lawrence Berkeley Laboratory is studying the impact of energy conservation in builings on indoor air quality.

Work at LBL on radon in buildings began in 1978. We have focused initially on the following tasks:

1. Review radon literature;
2. Enter radon information into LBL ventilation data base;
3. Develop a quick and sensitive technique for measuring the exhalation rate and diffusion length of radon in concrete and rock samples; and
4. Measure radon concentrations and ventilation rates in energy efficient buildings.

Interim results of this project will be presented.

The ultimate goals of the LBL indoor radon project are to:

1. Measure radon and radon daughter concentrations in conventional buildings and energy-efficient buildings;
2. Characterize sources of indoor radon;
3. Establish relationship between radon levels and building design parameters;
4. Evaluate control strategies; and
5. Estimate U.S. populations at risk to indoor radon in conventional buildings and proposed energy-efficient buildings.

THE EFFECTS OF VENTILATION ON RADON AND DAUGHTER ACTIVITIES

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The dependence of indoor potential alpha energy concentration on ventilation rate, source strenyth and outdoor Radon-222 concentration was studied usiny a computer program developed at the Center for Building Technology of the National Bureau of Standards. As an example, Figure 1 shows potential energy time courses for various source strengths; it is believed that 0.01 pCi/L min is a common value.

The model developed at NBS lends itself to simulation of source strenyths and ventilation rates changing at intervals of 1 or 2 hours. It can also be modified to account for settling out of particulates. Steady-state conditions have usually been assumed to study Radon-222 and daughter levels but are inappropriate because of changing air exchange rates and source strengths, and because they take several hours to reach at low air exchange rates.

A complete description of the program is presented in the forthcoming paper "Modeling of Radon-Daughter Concentrations in Ventilated Spaces" by T. Kusuda, S. Silberstein and P. E. McNall, Jr.

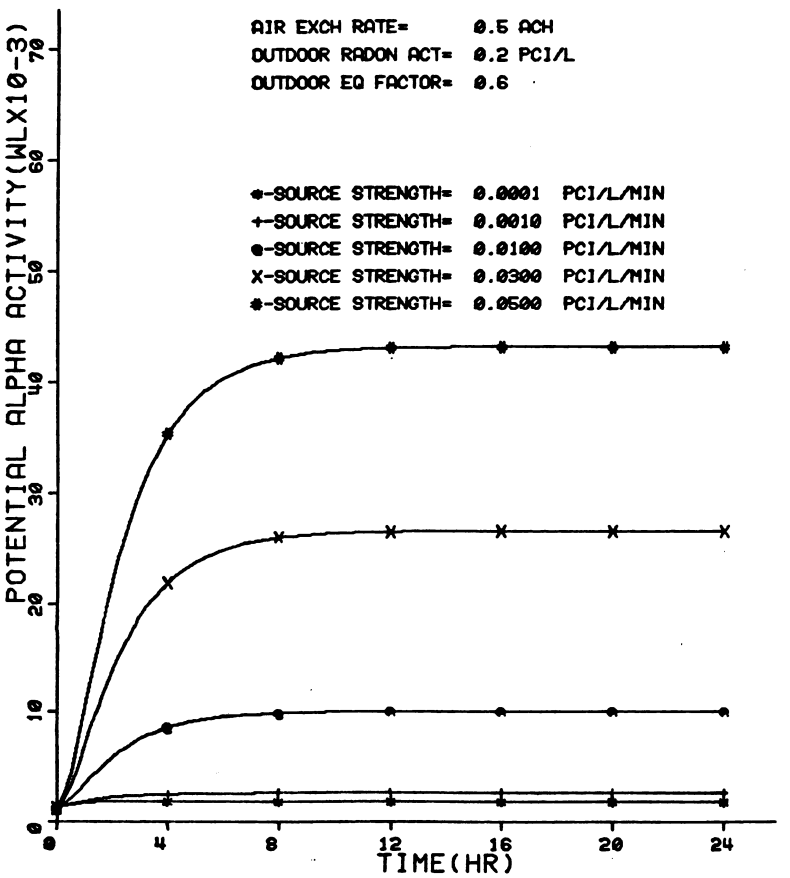

Figure 1. Potential alpha activity for various source strengths. Initial radionuclide concentrations equal outdoor levels.

* *

THE RADIOLOGICAL HEALTH ASPECTS OF SOLAR HOME HEATING

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Some active solar heating systems use a heat storage reservoir which consists of approximately 30 tons of granite. Air-cooled solar collectors transfer radiant energy to a bed of crushed granite, where the heated rock serves as a storage medium. The granite bed is usually in a basement of subterranean space; the rock is crushed to 1" - 1 1/2" diameter pieces. A conventional hot air heating system then extracts energy from the rock to maintain temperatures in the living space.

The hot granite also emanates ^{222}Rn. Preliminary calculations indicate that the granite bed may release 140,000 pCi per hour of ^{222}Rn. Allowing for losses of radon daughter products through leakage and filtration, an 1800-ft^2 home would maintain a level of 9 pCi per liter of ^{222}Rn and its daughter products or 76 cumulative working level months of exposure per year. This exceeds EPA's regulations for ^{222}Rn in homes. The zero-intercept, linear hypothesis model of the BEIR report suggests that continuous exposure of the US population at these levels would result in 26,000 to 56,000 cancer deaths per year.

Clearly, the potential health aspects of this energy technology have not been well examined. We suggest a three-part program of investigation:

1. Develop a complete analytical model which will examine the radiological influence of each design parameter of the solar heating system;
2. Inaugurate a measurement program which will test and verify the model; and
3. Select a solar heating design which will minimize radiological health impacts and also examine the tradeoff of radiation versus cost for a number of intermediate designs.

With only minor modifications this proposed model will be applicable to evaluation of the radiological health aspects of any type of solar heating design which employs masonry or stone heat storage media. This includes passive solar heating concepts, such as Trombe walls, masonry lined atria or greenhouses. In passive solar heating systems the radon emanation may be significantly lower; however, direct radiation from naturally occuring radionuclides in masonry materials is expected to be larger and may be as significant as the radon emanations from those active systems using rock storage beds. Consequently, the modifications to our model will include calculations of the direct radiation components.

The radiological health impacts of some solar home heating concepts or designs are potentially greater than other energy technologies. This is because radiation is released within the home rather than from a distant central power source. A single central power source may be more effectively and economically controlled than the dispersed sources which are inherent in a widespread deployment of solar space heating technology. Therefore, the radiological health implications of solar home heating must be factored into the research and development of this energy technology at the earliest possible stage.

* *

SOME DETERMINATIONS AT ARGONNE NATIONAL LABORATORY OF RADON IN HOUSES*

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The chance observation of a radon concentration of 26 $pCi \cdot L^{-1}$ in the bedroom air of a frame house has led to the discovery that such levels can arise as a result of emanation of radon from bare soil in a "crawl space" under part or all of the house. They are not a consequence of "technologically enhanced" radioactivity in building materials. In a total of 23 houses investigated, the air of ten showed concentrations of radon of 5 $pCi \cdot L^{-1}$ or more; of these, six had radon concentrations of 10 $pCi \cdot L^{-1}$ or more. It should also be mentioned that a concentration as low as 0.2 $pCi \cdot L^{-1}$ was observed in one of these houses. The presence or absence of plastic vapor barriers seems to be one important factor in determing the level, but certainly not the only one.

During July and August 1978, an Environmental Working Level Monitor that was returned to A.N.L. for repair and testing was used to determine the concentrations of radon daughters during a total of three periods, each of about 100 hours' duration, in two of the houses. Mean values of 0.008 WL and 0.023 WL were observed in the first, and of 0.008 WL for the second.

On the basis of such limited data it is obviously not possible to generalize on the average concentration of radon daughters in these houses. However, it is conceivable that the average concentration might be in the region of 0.01 WL; exposure at this level for a year (say of a small child) would result in a radiation dose equivalent of 2.6 rem to the bronchial epithelium (derived from 5 rem per WLM, for a 170-hour working month).

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*Work supported by the United States Department of Energy

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REPORT ON ACTIVITIES AT THE TECHNICAL UNIVERSITY OF DENMARK

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The radon-oriented work at our laboratory is presently concentrated around three topics:

1. EXHALATION OF RADON FROM BUILDING MATERIALS

As a part of the Danish Ministry of Commerce' energy saving projects a number of building materials are being checked for radon exhalation. The materials are being placed in closed containers and the build-up of radon activity in the containers measured as a function of time.

2. EFFECT OF IMPROVED INSULATION OF HOUSES ON INDOOR RADON LEVELS

As a part of the projects mentioned above the concentrations of radon and its daughter products are being measured in houses with very low ventilation rates. This will be done in newly constructed, so-called low-energy houses as well as in older houses and apartments, which have had their insulation improved. The radon measurements will be supplemented by the measurements of other indoor climatic parameters, such as temperature, humidity, ventilation rate and concentration of various (organic) vapors.

3. REMOVAL OF RADON DAUGHTERS BY ELECTRIC FIELDS

The effect of an electric field on the concentration of radon daughters in a closed room is being studied by keeping a wire or arrangement of wires at various positive and negative potentials with respect to ground. Preliminary measurements indicate that the working level may be lowered to about half of its undisturbed value by a single wire kept at a negative voltage of a few kV. The effect seems primarily to be due to a decrease in the concentrations of ^{214}Pb, ^{214}Bi and ^{214}Po, while the concentration of ^{218}Po remains almost constant, although ^{218}Po is the predominant element in the deposit on the wire. Work is planned to clear this problem as well as to study the relation between wire voltage and WL-lowering. Also the possible effect of a corona discharge on the depletion of the radon daughters will be studied.

* *

MEASUREMENTS OF AIRBORNE POLLUTANT CONCENTRATIONS INSIDE A WELL-INSULATED STRUCTURE WITH LOW VENTILATION RATE

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A series of pollutant concentration measurements has been made in a residential structure designed to reflect current concepts in energy conservation. The residence was constructed with ordinary building materials, but special attention was given to minimizing heat losses to the outside by air infiltration and surface convection. The house represents a typical eastern Tennessee dwelling: single family, two story, light frame, recirculated air, electric heat, and ventilated crawl space below the first floor. The structure is one of the three test houses (control house) included in the Tennessee Energy Conservation in Housing (TECH) project in Knoxville, Tennessee.

Measurements were made over a period of five days in September 1978. These included such determinations as concentration of indoor gaseous pollutants by mass spectrometry; indoor and outdoor radon concentration; indoor radon daughter concentration; radon flux through floors, walls and ground; airborne dust concentration; and ventilation rate in four rooms of the house. In addition to these measurements, samples of soil which were obtained from the crawl space beneath the house and from the lawn surrounding the house were analyzed for radionuclides in the uranium and thorium decay series. In order to make correlations between observed pollutant concentrations and meteorological conditions, data were obtained from existing TECH project instrumentation and included wind speed and direction, solar flux, temperature, and barometric pressure. Facilities and equipment used for this study included those of the Health and Safety Research Division's Off-Site Pollutant Measurements Group and the Monitoring Technology and Instrumentation Group.

Results of this study represent a baseline situation. In searching for nonradioactive airborne pollutants, no mass peaks above detection limit were observed. The detection limit for most mass peaks is 0.5 ppm. The low concentration of airborne pollutants is attributed to the fact that no one lives in the house; utility usage is simulated solely for purposes of economic studies. The mass spectrometer was useful for measuring the concentration of a tracer gas (CO_2) used to determine the structure's ventilation rate. This latter parameter was found to be dependent on external driving forces - notably wind speed. For example, whenever the wind speed was below about 10 kilometers per hour, the ventilation rate was approximately 0.22 air changes per hour (ach); however, as the wind speeds increased to about 15-17 $km \cdot h^{-1}$, the ventilation rate increased to 0.4 ach.

Continuous indoor radon measurements at four locations showed concentrations ranging from less than 0.1 $pCi \cdot L^{-1}$ to as high as 0.93 $pCi \cdot L^{-1}$. Most values fell within the 0.2 to 0.4 $pCi \cdot L^{-1}$ range. Simultaneously, continuous outdoor measurements ranged from 0.02 to 1.21 $pCi \cdot L^{-1}$ at two nearby locations. The indoor radon concentration was generally lower (20-50%) than the outdoor concentration. This reversal from the usual pattern may be attributed to the typically low ^{226}Ra concentration in building materials used in light frame structures of this type. Furthermore, the ventilated crawl space beneath the floor prevents soil-borne radon from readily diffusing into the structure. Other typical household sources of radon such as tapwater and natural gas result in limited contributions (none for natural gas) through simulated occupancy. Thus, most of the indoor radon originates from outdoor air entering the structure. For example, as the wind speed increased one day, the structure ventilation rate was observed to increase from 0.22 to 0.41 ach. The indoor radon concentration increased and approached that outside, but lagged the increase in exchange rate by three to four hours. This behavior was expected since outside air was the major source of radon. For structures coupled to the ground (i.e. slab on grade, or basement) constructed with materials known to bear ^{226}Ra (concrete, cinder block, brick, etc.), the reverse situation would be expected. That is, when the major source of indoor radon consists of that from construction materials and the ground, a reduction in ventilation rate would increase the indoor concentration. Results of radon daughter measurements at the four radon monitoring locations indicated that the degree of equilibrium between radon and its daughters ranged from 40 to 70% - a little less than would be expected at these air exchange rates.

The report contains all pertinent data collected during this study and includes tabular and graphical presentations of radiological and meteorological data as a function of time and location.

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*Consultant.

+Operated by Union Carbide Corporation under contract W-7405-eng-26 with the U.S. Department of Energy.

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RADON CONTROL IN HOUSING IN CANADA

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The Canadian government using the Atomic Energy Control Board as the executive agent of a Federal Provincial Task Force on Radioactivity has undertaken a program of decontamination and radon concentration reduction in a number of communities in Canada associated with the nuclear fuel cycle.

One town, Port Hope, Ontario, is the site of the Canadian facility where yellowcake is presently refined. Contamination in the town can be traced back over forty years from when this facility was used for radium extraction from pitchblende. Radon control in private, commercial and public buildings is primarily achieved by digging up and removing the contaminating soil from under and adjacent to those buildings where the radon/radon daughter concentration is found to be greater than the Task Force established criteria.

Three uranium mining towns, Elliot Lake and Bancroft in Ontario and Uranium City, Saskatchewan are also contaminated to some degree, in this case, by waste mine rock. However, the radon is mainly from natural sources and is transported underground by soil gas. All three towns are built on alluvial sands in proximity to uranium-bearing bedrock and our evidence suggests that there is a good

correlation between such porous soils and radon in structures. However we do not know the driving force that moves the gas. Clay will not permit long distance transportation of radon; unbroken bedrock does not emanate radon quickly enough to be a source for those buildings constructed directly on such rock formations.

Two remedial techniques have been developed:

(a) a sub-floor ventilation system; and

(b) the sealing of a foundation and the trapping of services which penetrate the foundation.

The latter approach is preferred as it causes minimum disruption to structures already in place. Application to new structures has been proven and the techniques can be made available to contractors as only the adaptation of standard building practices is involved.

* *

APPLICATION OF RADON STANDARDS TO NEW AND EXISTING HOUSING IN ELLIOT LAKE, ONTARIO

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Elliot Lake Project*
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A program to identify houses with radon daughter levels greater than acceptable (0.02 WL) and to take appropriate remedial action has been underway in Elliot Lake, Ontario, since mid 1977. The work is under the direction of DSMA/ACRES acting for the Atomic Energy Control Board (of Canada).

To date 671 houses have been surveyed and 117 have been identified as requiring remedial treatment. Of these, 74 have been brought to below the AECB criterion of 0.02 WL and work is proceeding on the remaining 43.

An exploration program extending over 6 months and culminating in a remedial demonstration program showed that the source of the radon was from the emanating fraction of naturally occurring radium in the soil and that the radon moved into the houses as a component of soil gas.

Investigation and testing techniques have been developed to locate the routes of entry of soil gas containing radon in finished and unfinished areas of all forms of house base construction, including poured concrete and block basement, slab on grade, and ventilated and unventilated crawl space.

Materials and methods for remedial treatment have been tested in the laboratory and in the field before being selected on a cost effective basis for specific application.

The National Building Code has been amended to provide that all new houses in Elliot Lake must be below 0.02 WL before occupancy is allowed. To assist contractors, three methods of construction have been developed; firstly a sub-floor mechanically-vented collector system which is incorporated into all mine owned homes; secondly, a passive system in which the design precludes the entry of radon into the houses and which is favoured by private purchasers; and thirdly a ventilated crawl space.

Direction of remedial work and the construction of new homes has shown that it is extremely difficult for an unsupervised contractor to construct a building without numerous unplanned openings. All failures to date have been caused by the inadequate levels of workmanship common to normal construction practices.

The order of magnitude of remedial and preventative construction work to date is:

Remedial Treatment	
Sealing of Single Route of Entry	$ 500.00
Fully Finished Basement including Apartment	$12,000.00
Average Cost of 74 Homes	$ 2,600.00
New Construction	
Under Floor System	$ 2,000.00
Sealed Basement	$ 500.00
Remedial Treatment on Unsupervised Construction	$ 2,400.00

*Dilworth, Secord, Meagher and Associates Limited, Elliot Lake Project in Consultation with Acres Consulting Services Limited.

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RADON STUDIES AT THE UNIVERSITY OF TEXAS

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Ed note: The speaker did not submit a summary for inclusion in this report. For further information contact him directly.

LEVELS OF RADON-222 IN MAINE AND NEW ENGLAND BUILDINGS ARISING FROM POTABLE WATER SUPPLIES

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During the past two years, the authors have been studying the gas ^{222}Rn in approximately 2000 groundwater supplies in Maine and 50 additional supplies elsewhere in New England. Levels of ^{222}Rn have been compared to physical, chemical, geological, and hydrological characteristics for 350 of the 2000 supplies. Levels of ^{222}Rn gas have been measured for several experiments in 10 homes and public buildings to determine the transfer from radon in water to air. Radon daughters have been determined to calculate the plate-out or loss from secular equilibrium and to enable dose calculations. Geographical distributions of ^{222}Rn levels in Maine private wells and utilities are given. Results of shower experiments for radon transfer into air and two day radon level measurements are given. The resulting variations of radon with time and water usage provide building ventilation time constants. Radon daughters in air are measured to be a factor of thirty lower than measured radon in air.

* *

CONTROL OF RADIOACTIVE HAZARDOUS WASTES PURSUANT TO THE RESOURCE CONSERVATION AND RECOVERY ACT (RCRA)

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Criteria and Standards Division
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Ed. note: The speaker did not submit a summary for inclusion in this report. For further information, contact him directly.

ROUNDTABLE DISCUSSION

Summary

This summary of the roundtable discussion attempts to accurately reflect many views and opinions that were expressed during the course of the discussion. It must be emphasized that it does not represent a consensus of the participants. Whenever possible, the summary maintains the chronological sequence of the discussion, but greater emphasis was placed on topically arranging the subject material. In all cases, contributions to the discussion are summarized or quoted without attributing them to a specific participant.

The roundtable discussion focused on four very broad, interrelated topics:

1. the existence of a potential health hazard and the scope of the problem;
2. sources of radon in buildings and characterization of those sources;
3. the adequacy of existing measurement methods; and
4. appropriate strategies and building technologies to reduce or control indoor radiation levels.

For each of these topics the participants were asked to address and, if possible, identify what is known, what are some of the suspected or possibly exogenous factors that may be involved, and what further research is needed.

* * * * *

Potential Health Hazard

The first topic focused on whether there indeed is a real problem arising from radiation exposure due to radon in buildings, and in particular, on whether the present concern over the health hazards is realistic. One participant noted that many of the existing conditions and sources of radon exposure have existed for many years, yet the regulatory agencies "seem to be on a red alert and gearing up" for treating these as problems. Is there really a significant problem, however, from the biological point of view? As an example, concern was voiced as to whether there is a public health hazard for residents of Maine who live in areas with naturally high concentrations of radon in their water supplies. In regard to this specific situation, it was reported that in looking at geographical areas in Maine with known concentrations of radon in water supplies and at cancer incidence in these areas (using National Cancer Institute data) some crude geographical correlations have been found. In some cases, such as lung cancer in women, the correlations are significant at very high significance levels ($p=0.001$). Less striking, but significant correlations have also been observed for lung cancer in men, female brain cancer, and male reproductive cancer. It was emphasized, however, that these results do not imply causality since many other factors could also cause cancer. Each of these factors would have to be considered before any conclusion as to the role of radon could be made.

Since available evidence does not exclude the possibility of a health hazard, the prudent view, as expressed by several of the participants, is to treat these situations as potential hazards with some associated risk. As a result, the estimated risk or risk estimator is of primary importance. One participant observed, however, that risk estimates are confounded by two very important considerations: (1) the cumulative level of exposure, and (2) exposure to other pollutants.

The first consideration involves the calculation of any cumulative unit of a radiation quantity, such as the Working Level Month (WLM), which is used as an indicator of the dose. Since one does not have an understanding of the cancer-induction mechanism, the last event that induces the cancer cannot be determined, and it is impossible to know when to stop the accumulation of dose in the calculation. Should one stop at death, at the time of diagnosis of the cancer, or should one try to account for some latent period? One participant indicated that all epidemiological data use WLM values that have been calculated up to the time of death. Risk estimates, therefore, are probably underestimated since some exposure occurs after the cancer was induced. It was noted that the risk estimates may increase if they are recalculated after uniformly subtracting the dose received during some assumed induction-latent period.

The second important consideration involves the problem of combined effects of radiation and exposure to other carcinogenic agents. Since exposure to radon daughters always occurs in the presence of other pollutants, we are never exposed to one risk at a time, and it is exceedingly difficult to extrapolate the results from one group to another. It was emphasized that it may be misleading to provide any risk estimate without further specifying the presence or absence of other agents. None of the current risk estimates considers combined effects. It was further noted that most of the lung-cancer mortality observed in uranium miners occurred in miners who smoked. The additional point was made that epidemiological data on bronchogenic cancers which were characteristic of miners in old uranium mines may not be applicable to populations exposed in non-mine environmental settings. Furthermore, there was a suggestion that some of the radon concentration data from uranium mines in this country and in Europe may be suspect. One participant said "I've been to many mines and as soon as I arrive I can hear the air pumps being turned on. By the time I get into the mine the working level is about 1 WL, but it was probably 10 WL before I got there."

Considerable discussion was devoted to the various extant estimates of lung cancer mortality resulting from exposure to radon daughters in residences. These mortality estimates range from a few percent to over 20% of all lung cancer deaths. Nearly all of them are derived in part using the risk estimators of the UNSCEAR* and BEIR** reports along with other assumptions about activity levels, occupancy and intake factors. Several conflicting arguments were made regarding the validity of some of the mortality estimates. In all cases, these conflicts involved the underlying assumptions employed in the calculations. It was observed, for example, that most indoor levels are five to ten times smaller than the value of 0.02 WL assumed in one estimate. Another participant concluded that one cannot use current exposure levels to predict current incidence rates since houses were leakier in the past than now, and lung cancers have latent periods of 20 to 30 years. Therefore past levels should be used to predict current rates, and present levels for future rates. Additionally, the assumed equilibrium factors for the daughter activities in houses may not be correct. The daughters ^{214}Pb, ^{214}Bi, and ^{214}Po contribute significantly to the dose, and their concentrations may differ by a factor of ten in a residence. This effect alone introduces considerable uncertainty to the mortality estimates. Perhaps the most telling point was made by the participant who suggested that an entire meeting "lasting a week or even a month" could be devoted exclusively to the topic of "what are the best values [of the risk estimators] to use."

An entirely different aspect of this question as to the existence of a problem was subsequently raised in a later part of the discussion by a participant who suggested that the true scope of

*United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), "Sources and Effects of Ionizing Radiation", 1977 Report to the General Assembly, United Nations (1977).

**"The Effects on Populations of Exposure to low Levels of Ionizing Radiation", National Academy of Sciences, Advisory Committee on the Biological Effects of Ionizing Radiation (1972).

the problem will remain unknown until more field studies are conducted. Only then will we be able to say what the effects are, and be able to derive some maximum estimate of the risk. He then expressed the hope that we will be able to evaluate these risks on an equivalent basis with other radiation risks. "From the public health point of view, in the best of all possible worlds, we would hopefully be able to spend our money to reduce radiation exposure in ways which will do the most good." That is, in ways that will result in the quickest and largest reduction for the money spent to maximize the risk reduction per dollar. He lamented that political factors and misinformed public perceptions of the various risks play a more important role than the magnitude of the risk. As an example he offered the case of natural gas, which is a source of radon in buildings when used in unvented space heaters and appliances. The Environmental Protection Agency (EPA) six years ago estimated that this source results in a population dose commitment of 2.7 million person-rem per year, resulting in an estimated 95 lung cancer fatalities per year. He suggested that this has been completely ignored while billions of dollars are spent in the nuclear power industry to reduce dose commitments as low as 0.05 million person-rem per year. A participant from EPA concurred and stated that the nuclear power industry has indeed spent on the order of hundreds of thousands of dollars per life saved for public exposures and much larger sums for occupational exposures. "One of the things that is encouraging about this conference.....is that we are now focusing on a radiation problem where the returns per dollar spent are much, much greater." He provided, as an example, the implications of the EPA recommendations for Florida [subsequently published in Federal register 44, 38774-70 (July 2, 1979)]. It has been calculated that exposure to 0.02 WL in an average house results in a 2% risk of lung cancer per individual for lifetime occupancy. If remedial measures costing approximately $1000 per house are made, it is estimated that we will be spending at a rate of $12,000 per health effect averted (assuming 4 occupants per house). This may be contrasted to the hundreds of thousands or millions of dollars spend in the nuclear industry. He suggested that by focusing on these areas great returns can be achieved, and will result in more equity in the treatment of radiation risks.

The issue of equity was addressed by another participant who noted that although this dispassionate approach may be intellectually satisfying and the altruistic ideal, it is not realistic in a democratic society. We stress the risks the public or body politic wishes to stress. "If the public is more concerned about a microrad from a nuclear power plant than a millirad from radon, then we are going to be worried about nuclear power plants."

Subsequent to the conclusion of the roundtable discussion, the question of whether there is a problem was summarized by a participant who concluded that "there is indeed a problem and that it is quite simply this - what is the average radiation dose from radon daughters to the lung experienced by the population of the U.S.A., how does it vary with type and location of house, and what can be predicted about future trends of that dose as houses are built with less ventilation, or with additional sources of radon (underground houses, houses in caves, houses with thermal storage in rock, etc.)? This is the real issue and it is an extremely important one in view of the great public concern about tiny and probably inconsequential increments of radiation dose from nuclear power, either as a result of normal operation or of accidental releases (e.g., Three Mile Island)."

* * * * *

Source Characterization

The second major topic of discussion focused on the sources of radon in buildings and characterization of these sources. It was noted many times during the meeting that there are a myriad of source variables, such as the radioactivity content of the source, the porosity and moisture content of materials, meteorological effects, temporal changes, and air exchange rates. The

participants were asked to explicitly address these source variables from the perspective of whether we know how to characterize the sources of radon in buildings, what is known, what is unknown, and what are the research needs.

The measurement of emanation rates* from various building materials was immediately identified as one area in which further research would be highly useful. It was noted that radon escapes very readily from some materials, but not from others, such as phosphate slag in Butte, Montana which seems be be highly vitrified. One participant indicated that a crucial variable for the emanation rate is the water content of the material. Another participant reported that preliminary results from work (being done under contract for the Mine Safety and Health Administration) to characterize the effects of moisture content on emanation from uranium ore indicates that there is a specific water content that results in maximum emanation. The emanation rate is lower at both very high and very low water concentrations.

A significant point was made by another participant who indicated that if the important parameter is the amount of radon that is released from materials, then an emanation rate measurement standard may be more appropriate than standards for the radioactivity content of the material. He said that the measurement of emanation rates has received very little attention in the past, and suggested that we need to give more consideration to methods for measuring emanation rates, to standardizing the methods, and to establishing mechanisms for measurement intercomparisons between laboratories.

Another participant suggested that in some cases present measurement methods are probably adequate. He reported that, in a twenty-house study in the New York-New Jersey area done by the Environmental Measurements Laboratory (EML),the annual mean radon concentration could be accounted for by considering the radon flux from the cellar floor, radon concentration in water, and an inferred average ventilation rate. Measurement data for these variables were subjected to multiple regression analyses, and the results were then used to obtain calculated values of the average annual working level which were in agreement with directly determined levels. The conclusion is that the results are "perhaps better than we have a right to expect", and that the "measurements were fairly good."

The adequacy of existing measurement methods, with respect to emanation rates, was also addressed. It was reported that the Technical University of Denmark has characterized emanation rates for several materials. This participant indicated that studies show the material should be characterized in terms of the emanation rate per unit surface area in some cases while in other cases it is more relevant to describe the material in terms of the emanation rate per unit mass. To a large extent, it depends on the porosity and permeability of the material. He also suggested that it is completely possible with existing techniques to characterize emanation rates of materials under various conditions (humidity, pressure, etc.). "It's not so much a question of technique, it's more of just doing the work."

The issue was raised whether these measurements, to be of particular use, should be made on building materials when they are in place or as they are actually used (e.g. on wallboards after they are painted). Similarly, the emanation into a room will depend on the amount of thermal insulation and on the presence of a vapor barrier. Some possible problems with these in situ measurements were considered. The results may be confounded by ground-soil radon or by other sources in the building. It was indicated that one would certainly want to characterize the site before initiating such in situ building material measurements. An alternative approach is to characterize the basic building materials, from which you can then usually establish an upper limit on what this material will contribute to a

*In response to a specific question about the use of activated charcoal for the measurement of radon emanation by the charcoal canister method, a participant reported that coconut charcoal is the best choice.

given construction.

The relative importance of the various sources of radon was also considered. It was reported that in buildings on reclaimed phosphate lands in Florida, some radon comes from building materials, but most originates from the soil outside the building. Similarly, Canadian studies indicate that most of the radon originates from soil leakage. These areas in Canada use lake water and do not have the problems associated with high-radon water supplies that might be found in Maine. In the work at Elliot Lake, Ontario [see p. 66], it was found that the major source of radon in buildings was not concrete or other building materials, but rather was soil gas containing radon which entered by discontinuities in the construction, floor cracks, drains with improper traps, etc. In 74 houses in Elliot Lake, radon infiltration was significantly reduced by blocking the routes of entry of soil gas into basements. In the EML study referred to earlier, emanation rates were measured over floor cracks in older houses as well as over intact concrete floors in newer houses. Although the ^{226}Ra concentration in the soil around the houses is fairly uniform at approximately 1 $pCi \cdot g^{-1}$, and all building codes in the New York-New Jersey area require a 4-inch thick concrete floor in the basements, a 10-fold range in the average radon flux from the floors was found. The causes of this variation, part but not all of which can be explained by cracks, are unknown. It was suggested that these wide variations, as well as similar 100-fold ones seen in houses with unvented crawl spaces, indicate that the rate depends on the type of soil. It was also pointed out that soil type is only one of many factors. Depending on conditions above the soil, 100-fold changes in a 12-hour period can be observed. Soil is a major source in many cases, but this is not necessarily universal. Some studies in Denmark, for example, indicated that radon originated primarily from building materials. Indoor radon concentrations could be calculated from exhalation rates of walls and floors. Significantly, no differences in the rates between floors in contact with soils and walls not facing soils were found. In the course of the discussion it became apparent that the source of radon is very situational and site specific, and that characterization will depend on the specific type of source, viz., water, building materials, soil.

Earlier in the discussion, it was implied that we know how to characterize the sources, and that we only need to go out and do the work. This was viewed somewhat skeptically, however, by several participants. One asked, for example, if we know how to predict what the WL in a structure built on reclaimed phosphate lands in Florida will be before it is built. Past efforts have not been very successful, and it was asked if we can now do better by considering additional parameters such as land class [as described in one of the Brief Reports, p. 56]. It was clarified that land class is not an aid in predicting. Land classification is just a manifestation of the soil radium content and, perhaps as a second order effect, the radon emanation rate of the soil. The suggestion was made that we need to focus on soil radium content and perhaps other parameters, including simpler ways of measuring both the lateral and vertical distribution of radium in lands. It was pointed out that we should not completely judge things on the basis of the situation in Florida. Present land reclamation techniques are such that the radium concentration varies greatly both vertically and horizontally within very short distances. Soil radium concentrations can differ by a factor of 10 within a few feet. One of the major problems that must be addressed is finding appropriate techniques for averaging over distance and depth. Measurements of external gamma-radiation exposure rates, which in effect integrate over large areas, may be a better predictor than anything else because of these drastic variations. In Canada, similar gamma-radiation surveys are conducted to detect site anomalies. Obviously, one does not want to build a housing development on a radon anomaly.

The question of prediction appeared to be very important to a number of participants. One of them, referring to the EML study, found it interesting that the WL could be predicted on the basis of emanation rate measurements. He suggested

that it is easier to measure radon concentrations in air than either emanation rates or radon-daughter concentrations in air. He proposed, however, that existing WL monitors [e.g., described by Keefe, et al., p. 54] can be used to obtain average radon-daughter concentrations. Using data collected in an Argonne National Laboratory study [see p. 63], he demonstrated that one could obtain $^{218}Po/^{214}Bi$ and $^{214}Pb/^{214}Bi$ concentration ratios under different conditions with available instrumentation. For example, measurements indicated that when an air conditioner fan was used continuously, the ratios were reduced by half compared to when the fan was off or used only in conjunction with the compressor. He concluded that it is possible, irrespective of the source of radon, to obtain valuable information on the average radon-daughter concentrations and thereby predict the WL if a sufficient number of measurements, under a variety of conditions, are made with these existing instruments.

The discussion shifted to a related, but slightly different aspect when another participant asked if it is the respirable fraction of the radon daughters that is hazardous. If it is, could the measurement problem and the question of prediction be simplified by focusing on known particle-size distributions and measuring the activity on just the respirable fraction? This was viewed as an oversimplification since the daughters, in the decay sequence $^{218}Po \rightarrow {}^{214}Pb \rightarrow {}^{214}Bi \rightarrow$, can attach, unattach, and reattach to any size particles in air. It was also noted that the radon daughters are almost always respirable in the conventional sense of particle-size definitions. For lung dose assessments, the unattached or uncombined fraction of the radon daughters, and not the respirable fraction, is the critical size factor. Unattached fractions, however, are not usually measured. It was further suggested that air filtration and recirculation results in a reduction of the ^{214}Pb and ^{214}Bi daughters because the unattached fraction of their precursor ^{218}Po deposits on filters and fan blades and remains attached to these surfaces. The ^{218}Po is not similarly decreased since it is continually replenished by decay of ^{222}Rn in the air.

* * * * *

Measurement Needs

Identification of specific measurement needs was the major focus of the next topic of discussion. It was suggested that this has to be considered from the perspective of having differing needs, depending on whether one is interested in broad-based surveys or monitoring of buildings, or if one is trying to understand the problem or study the mechanisms and systems involved, or if the measurements are being used to help establish standards or compliance criteria. Survey measurement needs appeared to be the area of most immediate and greatest concern.

It was noted that we are increasingly forced to rely on State radiation control offices to make the necessary measurements. They have primary responsibility, but they are "already pushed to the limit" and do not have adequate resources to make complicated measurements. Florida, for example, surveyed 100 houses over several years and selected one measurement technique--track etch film--mainly because it is a simple, relatively inexpensive, passive method. The number of houses we may be faced to deal with is not encouraging. For example, the TVA phosphate-production facility at Muscle Shoals produced, over many decades, huge quantities of phosphate slag that was incorporated into concrete building blocks. It is estimated that there are 100,000 to 200,000 houses in four states which were constructued with these blocks. The State of Alabama is conducting a small pilot study of these houses, and it appears that there may be a problem requiring remedial action, but we have no idea on how to survey a population of houses of this magnitude. It was emphasized that it certainly can not be accomplished with conventional air-sampling methods. What is needed is a simple means of integrating annual exposure. Another participant referred to the previously reported [see p. 63] chance observation of elevated indoor radon-daughter levels which were not a consequence of "technological enhancement," and

rhetorically asked if it is reasonable to survey all homes in the United States.

The implied need for more inexpensive instruments was questioned by another participant. His view was that labor is expensive, and it contributes to the high cost of measurements. Therefore, automated instruments are needed instead. The initial cost of an automated instrument is admittedly higher, but the final cost of the measurements will be much lower.

It was suggested that we do not yet know how to optimize our measurements. We know that single, short-term measurements are inadequate for characterizing a house. One participant felt reasonably confident in week-long integral measurements taken quarterly or more frequently over a year. The optimum is probably somewhere between these two extremes, but we do not know where it is. Another participant indicated that if a house has very low or high levels,then many measurements are needed because of the inherent statistical difficulties. In addition, another important factor is that occupants become annoyed by the intrusions of frequent measurements. One must reconcile this practical problem when trying to survey an occupied house. It was reported that in Canada a comprise of 13 short-term measurements over a three-week interval is made.

The suitability of the track etch technique was discussed in considerable detail. One participant felt that although the measurement costs are much less than other instrumental methods, his experience was that this method is too imprecise and unreliable. He added that if the technique could be refined it would be very useful for surveying thousands of houses. Another participant indicated that some of the early surveys made with this technique were done during the initial phases of applying it to measurements at environmental levels. He indicated that there have been significant improvements in precision, sensitivity and discriminatory ability of the material. Track-etch materials are simple devices that allow one to make integral measurements in many houses at a cost of approximately $10 per house. Track-etch cups can be employed with a variety of membranes to achieve different hold-up times, and can also be used with a simple filter barrier to exclude the radon daughters. One participant encouraged further development, and hoped that the acceptability of this technique can be demonstrated for it appears to be the only economical mass screening device on the near horizon.

The length of time needed to obtain measurements was also raised as another practical problem. If, for example, a regulation specifies an upper limit of 0.02 WL in a house, does the builder constructing to this specification have to wait a year to obtain the results? Further, if remedial action is needed, does one have to wait a year to learn if the corrections were adequate? It was suggested that in many of the cases construction errors are so gross that they show up very quickly. This is not always the case, however, particularly in the "grey area" where you may find individual measurements in the range of 0.01 to 0.03 WL and where the average may very well be above 0.02 WL. The ability to predict in a short period of time whether a house meets compliance criteria was suggested as an area requiring further research. Another approach may be to characterize the conditions which will allow one to predict the working level. In this way, one could build the house in the right way to satisfy the conditions without making any measurements. It was pointed out, however, that this may be slightly visionary. Based on experiences in Canada, major flaws can occur in houses built under even the most demanding construction specifications. At Elliot Lake, for example, one house built under direct supervision passed inspection while a similar house built by the same builder but without supervision failed. After investigation, to the dismay of the builder, it was found that its failure was partly due to the absence of a caulking strip at the floor-wall joint for a stretch of nearly 5 meters!

* * * * *

Control Technologies

The last topic of discussion focused on the adequacy of existing control technologies, and on

how concern for indoor radon pollution may influence building practices.

Primary emphasis was centered on methods to reduce or control the WL in buildings. One participant asked if the only practical method was increasing or maintaining high air-exchange rates. If so, it has a large economic penalty. It was noted that buildings consume 30% of the energy used in the United States, and that ventilation, heating, cooling and conditioning of ventilation air is 20 to 30% of that. Therefore, ventilation currently comprises 5 to 10% of total U.S. energy use. To reduce these energy-use costs, it may be worthwhile to assume considerable front-end capital investment and construction costs to reduce the WL by other means. Another participant suggested that the role of ventilation may be overemphasized. He reported that in tests at Elliot Lake on both electric and oil heated buildings, measurements of ventilation rates and radon and radon-daughter concentrations indicated that: 1) fluctuations in radon concentrations and working levels are mainly caused by variations in the radon supply rate into the house rather than by variations in the ventilation rate; and (2) equilibrium fractions and radon-daughter concentration ratios are almost independent of ventilation rate and mainly depend on the plate-out rate of ^{218}Po. It was pointed out that this implies that the equilibrium fraction is the same inside and outside the building, and will not be affected by a factor of two or three change in the ventilation rate in the absence of recirculation.

Air cleaning or filtering was considered as another possible remedy. It became apparent, however, that this may not be effective. Electrostatic precipitators or other electronic air cleaners will not work unless the particles can be charged. Furthermore, the first decay product, ^{218}Po, is continually replenished in the air by decay of the radon, and the radon itself cannot be removed from air by filters or by any other practical means. The only known ways are to freeze it (e.g., with liquid air) or to trap it in charcoal. Because of the maintenance problems, there does not currently appear to be any satisfactory mechanism for maintaining charcoal or media filters in a residence.

The suggestion was made that we may be looking too parochially at the control of radon. Certainly efforts should be made to control the sources of radon since we have seen anomalies in some conventional residences, but there are also many other indoor contaminants. For example, formaldehyde originating from "chip board" glues and other building materials has been found in tight trailers at concentrations exceeding occupational standards. The number of potential contaminants is great, and a solution which consists of eliminating the source of only one - radon- and then tightening the house is not adequate. Unless all contaminants are known and controlled by other methods, which is not probable, we must supply adequate ventilation. It was recommended that this could be done by heat exchangers. Another participant agreed that this is a solution, but one that bears an economic penalty. The efficiency of simple regenerators is of the order of 60%, so about one-third of the energy would still be wasted. This is a clear case of the trade-off between energy and equipment costs and ventilation rate. It was suggested that an integral heat exchanger system may be cost effective in American homes that already utilize central forced-air heating systems. One participant thought that any control measure must be permanent since the problem remains with the building over its lifetime. One needs either a passive control measure or one that remains an integral part of the building's services which cannot be turned off or tampered with.

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The roundtable discussion clearly demonstrated that much is known about radon in buildings. It also demonstrated, however, that there are many uncertainties and outstanding needs, and that significant questions must still be resolved.

Various risk estimators, primarily derived from data on radiation exposure in uranium mines, are available to quantify the risk of lung cancer due to inhalation of radon-decay products. We do not know, however, if the risk estimators are

valid for low-dose environmental exposures. It is not known, for example, if the extrapolations to lower doses are legitimate, or if the etiology and types of cancer are applicable in non-mine environmental settings.

Many sources of radon can be characterized with known variables using existing methods. In most situations, however, we do not know how to systematically treat the source characterization in ways which will allow us to make predictions. For example, given a specific building type and a known concentration of radon in the building's water supply, or the radium content in the soil surrounding the foundation, we can not predict the average radon-daughter concentrations in the building air, or more importantly, predict the occupants' average radiation dose to the lung. In cases such as these, it is not known if the variables we are presently measuring are appropriate and most significant for the problem.

Suitable measurement methods exist for many applications. Notable exceptions include reliable, inexpensive screening methods to evaluate undeveloped lands bearing relatively high levels of radium and to survey large numbers of existing buildings. The adequacy of methods which have been suggested as showing promise - such as track-etch techniques - needs to be unequivocally demonstrated. In addition, there is a great need for measurement standards and interlaboratory comparison programs that can demonstrate the adequacy and reliability of measurements.

A number of remedial and preventative construction techniques - mainly removal of contaminating soil, installation of subfloor ventilation systems, and sealing of foundations and services which penetrate the foundation - have been successful in reducing and controlling radon and radon-daughter concentrations in buildings. These have for the most part been applied in situations where the major source of radon was from infiltration of soil gas into the building. It is not apparent, however, that these techniques are applicable for other sources of radon such as emanation from building materials or release from water. In cases where control relies exclusively on supplying adequate ventilation, we do not know how to provide efficacious methods which are consistent with the need for reducing energy consumption.

It is hoped that this discussion has in some small way contributed to a better understanding of the problems, and provided a stimulus for further experimental and theoretical work on the subject.

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This is the proceedings of a Roundtable Discussion of Radon in Buildings held June 15, 1979 at the National Bureau of Standards in Gaithersburg, Maryland. The meeting brought together a number of participants with diverse interdisciplinary interest in radiation protection, radiation measurement and building technology, provided a forum to exchange information, and drew attention to some of the problems and research needs associated with radiation exposure due to radon in buildings. Emphasis was placed on (1) the characterization of the sources and pathways of radon in buildings; (2) the biological and health effects; (3) measurement considerations; and (4) strategies and control technologies to minimize indoor radiation exposure.

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