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RADON DOSIMETRY: A REVIEW OF RADON AND RADON DAUGHTER EXPOSURE CONDITIONS IN DWELLINGS AND OTHER STRUCTURES

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§Health and Safety Research Division.

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ACKNOWLEDGEMENTS

The authors would like to express their thanks to those individuals who kindly supplied reprints and reports which were not readily available previously. Their willingness to supply this information contributed significantly to the analysis.
ABSTRACT

Within the past few years several situations have been brought to light which indicate an increased radiation exposure of certain segments of the general population caused by human activities. The most widely publicized activities are those associated with the mining and milling of uranium in the western United States, the phosphate industry in Florida, and those potential problems represented by former Manhattan Engineer District sites. One of the primary problems involves exposure to radon and radon daughters which are released from large waste piles or, in some cases, evolve from backfill and construction materials used in homes, schools, and other buildings.

This report presents a review of the available data on radon and radon-daughter concentrations in dwellings and other structures. The primary objectives were to compile and tabulate pertinent radon exposure data and to prepare a statistical summary of the data which will be useful in the prediction of normal levels of radon and radon-daughter concentrations in these structures. In addition, other parameters associated with radon exposure conditions are presented and discussed.
1. INTRODUCTION

Public awareness of exposures to ionizing radiation has focused primarily on nuclear power generation of electrical energy. However, more recently there has been increased concern about exposures associated with the enhanced natural radiation environment. Several situations have been brought to light within the past few years which indicate an increased exposure of certain segments of the general population caused by human activities. These include, but are not limited to, commercial activities related to the nuclear fuel cycle. The most widely publicized are those associated with the mining and milling of uranium in the western U.S., the phosphate industry in Florida, and those potential problems represented by former Manhattan Engineer District (MED) sites.

Large waste piles from the uranium and phosphate industries emit gaseous radionuclides which are sources of exposure for those individuals living downwind. In addition, at a few sites with unique hydrologic conditions increased levels of naturally occurring radionuclides have been found in streams and drinking water supplies located near these waste areas. The problem arises, however, with the use of these wastes as backfill and construction materials in homes, schools, and other buildings which are frequently occupied.

The absorbed dose resulting from exposure to radon and radon daughters involves two interrelated areas: (1) the behavior of radon and radon daughters in the atmosphere to which the individual is exposed and (2) the deposition, and subsequent energy release as the inhaled daughters undergo radioactive decay. Many years of research have been invested in radon
dosimetry related to uranium mining situations. However, the extrapolation of these results to individuals in the general population exposed to lower radon daughter concentrations and atmospheres of different compositions remains questionable. It is reasonable to expect that the absorbed dose due to radon-222 and its daughters can be expressed as a single numerical value applicable under a variety of exposure conditions (Johnson, Hardin, and Nelson, 1975).

This report presents a review of the available data on radon and radon daughter concentrations in dwellings and other structures. In addition, other parameters associated with radon exposure conditions will be discussed. The primary objectives were to compile and tabulate pertinent radon exposure data and to prepare a statistical summary of the data which will be useful in the prediction of normal levels of radon and radon daughter concentrations in these structures. The ability to predict these normal levels in advance should facilitate meaningful evaluations of the levels and associated exposures.

2. RADON AND RADON PROGENY EXPOSURE CONDITIONS IN DWELLINGS

2.1 GENERAL APPROACH

This literature review included a large number of papers and reports published within the last fifteen years. Since the radon daughter problem has been studied more extensively in recent years, the evaluation was concentrated on information published in the last five to six years. Many of the papers described methods of measuring radon and radon daughter concentrations in various environments. The purpose of evaluating these
papers, in addition to others which were more directly relevant, was two-fold. First, this allowed a better understanding of the sensitivity of the particular detection method and the accuracy of the data obtained. Secondly, many of the papers included limited data obtained with the detection system, during checkout and testing, in dwellings and other structures.

The next step in this particular phase of the review involved a tabulation of those data relevant to the evaluation. The data were presented in several ways. Some authors reported the concentration of radon-222 measured, usually in units of $\mu$Ci/L or pCi/m$^3$. Other authors presented their measurements in terms of the working-level (WL)*. In many cases, the concentration ratios of RaA, RaB, RaC, or RaC' were not published. Finally, some authors reported measured levels of radon daughters, usually in units of pCi/L.

In this review, radon concentrations were accumulated in units of pCi/L while radon progeny concentrations were in units of the working level.

Due to the wide variety of data available, several groupings were established. First, all data from the United States and Canada were grouped together. These two countries share four of the seven major land regions of the North American continent. Therefore, the separation of the U.S. and Canadian data did not seem justifiable. This large group of data was subdivided further into three categories. These were:

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*WL is any combination of short-lived radon daughter products in 1 liter of air that will result in the ultimate emission of $1.3 \times 10^5$ MeV of potential alpha energy.*
(1) residential structures built on unaltered land, (2) basements of residential structures built on unaltered land, and (3) residential structures built on altered land. Category 1 included dwellings in regions of Canada with elevated levels of the uranium series elements in the soil. However, the land was not altered by man's technological activities and, thus, represented essentially a natural exposure situation. The primary source of data from dwellings constructed on altered land came from areas in the western U. S. where tailings materials had been used and also from the phosphate areas of Florida. Where it was known that concentrations of radium in soil were greater than 5 pCi/gm, radon and radon progeny data were not included. Radon and radon progeny data were included in a grouping only where radium soil concentrations were less than 5 pCi/gm.

For each of the data groupings mentioned above, radon (or radon progeny) concentrations were separated into seven or more concentration levels. All data which comprised a particular group (e.g., residential structures built on unaltered land) were pooled even though the data may have been obtained by different investigators using different techniques. At the completion of this pooling, the number of observations in each "concentration bin" was divided by the total number of observations recorded. These data were used to produce cumulative distributions which will be discussed in subsequent sections of this report.

All other papers published by foreign authors, i.e., from countries not part of the North American continent, were grouped in a similar fashion. Essentially all of these papers were the results of research performed in Europe. Although the data for North America are of primary interest here, these "foreign" results will be discussed also.
2.2 RADON CONCENTRATIONS IN DWELLINGS

Measurements of radon concentrations on the first floor (main floor) of residential structures in the United States and Canada are presented in Table 1. All measurements were pooled and a frequency distribution for the entire set of data was obtained. The data reported by McGregor (McGregor et al., 1980) were not included because approximately 60% of the data points were grouped in the range 0.0-0.5 pCi/L which prevented adequate evaluation of the distribution of this data. These data, in the form of a lognormal distribution, are also shown in Fig. 1. The mean radon concentration from Fig. 1 is 2.5 pCi/L with a geometric standard deviation of 4.8.

Radon concentrations measured in basements located in the U. S. and Canada are summarized in Table 2. As before, the data were pooled and plotted in the form of a lognormal distribution (Fig. 2). From these data, the mean radon concentration in basements is 7.1 pCi/L with a geometric standard deviation of 3.5.

Measurements of radon concentrations in dwellings constructed on altered land have not been widespread. Roessler, Wethington and Bolch (1978) made only a few measurements in the Florida phosphate area. The mean value of their measurements was 4.02 ± 6.29 pCi/L. The range of measurements was 0.4 to 23.8 pCi/L.

There is a large volume of data available from foreign countries but these data are not directly relevant to the evaluation of radon and radon progeny levels in North America dwellings. Therefore, the foreign data were not analyzed in detail. A summary of the collected data is given in Table 3. Note that the data presented in this table bracket
<table>
<thead>
<tr>
<th>Reference</th>
<th>Country, state</th>
<th>Mean (pCi/L)</th>
<th>Range (pCi/L)</th>
<th>Comments</th>
</tr>
</thead>
<tbody>
<tr>
<td>Keith Consulting (1978)</td>
<td>Canada</td>
<td>6.7 ± 8.3</td>
<td>0.056 - 44.2</td>
<td>223 measurements at Uranium City, Saskatchewan</td>
</tr>
<tr>
<td>Breslin, Knutson, and George (1978)</td>
<td>New Jersey, New York</td>
<td>1.0 ± 0.8</td>
<td>0.25 - 3.1</td>
<td>15 measurements in selected homes</td>
</tr>
<tr>
<td>Dillworth, Secord, Meagher, and Associates</td>
<td>Canada</td>
<td>6.3 ± 7.7</td>
<td>0.1 - 33.0</td>
<td>20 measurements in Elliot Lake, Ontario</td>
</tr>
<tr>
<td>Limited (1978)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Roessler, Wethington, and Bolch (1978)</td>
<td>Florida</td>
<td>1.2 ± 1.3</td>
<td>0.2 - 4.1</td>
<td>15 measurements on unaltered land in two counties</td>
</tr>
<tr>
<td>Caruthers and Waltner (1973)</td>
<td>North Carolina</td>
<td>0.46 ± 0.36</td>
<td>0.005 - 1.6</td>
<td>25 measurements in 11 homes, construction unknown</td>
</tr>
</tbody>
</table>

From Figure 1: Mean concentration = 2.5 pCi/L; geometric standard deviation = 4.8 pCi/L.
Fig. 1. Radon concentrations in residential structures in the United States and Canada — data from main floor only.
Table 2. Radon concentrations in residential dwellings, basements only — United States and Canada data

<table>
<thead>
<tr>
<th>Reference</th>
<th>Country, state</th>
<th>Mean (pCi/L)</th>
<th>Range (pCi/L)</th>
<th>Comments</th>
</tr>
</thead>
<tbody>
<tr>
<td>Keith Consulting (1978)</td>
<td>Canada</td>
<td>19.3 ± 54.0</td>
<td>0.02 — 455.14</td>
<td>255 measurements at Uranium City, Saskatchewan</td>
</tr>
<tr>
<td>Breslin, Knutson, and George (1973)</td>
<td>New York, New Jersey</td>
<td>1.79 ± 1.19</td>
<td>0.39 — 4.4</td>
<td>16 measurements in selected homes</td>
</tr>
<tr>
<td>George and Breslin (1978)</td>
<td>New York, New Jersey</td>
<td>0.96 ± 0.38</td>
<td>0.45 — 1.6</td>
<td>7 measurements during a test of cumulative environmental radon monitor</td>
</tr>
<tr>
<td>George, Breslin and Guggenheim (1976)</td>
<td>New York</td>
<td>6.0 ± 1.5</td>
<td>3.6 — 7.8</td>
<td>11 measurements on New York City office building</td>
</tr>
<tr>
<td>Spitz and Wrenn (1974)</td>
<td>Colorado</td>
<td>44.6 ± 11.6</td>
<td>30.8 — 58.4</td>
<td>6 measurements in Grand Junction, Colorado</td>
</tr>
<tr>
<td>McGregor et al.* (1980)</td>
<td>Canada</td>
<td>-</td>
<td>0.14 — 0.88</td>
<td>9113 measurements in 13 Canadian cities</td>
</tr>
</tbody>
</table>

From Figure 3: Mean concentration — 7.1 pCi/L; geometric standard deviation — 3.5 pCi/L.

*Not included in Figure 3 (see text for discussion).
Fig. 2. Radon concentrations in residential structures in the United States and Canada — data from basement only.
Table 3. Summary of foreign data on radon levels in dwellings and other structures

<table>
<thead>
<tr>
<th>Reference</th>
<th>Country</th>
<th>Mean (pCi/L)</th>
<th>Range (pCi/L)</th>
<th>Comments</th>
</tr>
</thead>
<tbody>
<tr>
<td>Steinhausler (1975)</td>
<td>Austria</td>
<td>1.02 ± 0.3</td>
<td>0.15 - 4.7</td>
<td>Data for 11 locations, brick, stone, concrete construction</td>
</tr>
<tr>
<td>Jonassen (1975)</td>
<td>Denmark</td>
<td>—</td>
<td>3 - 7</td>
<td>Actually basement rooms of concrete</td>
</tr>
<tr>
<td>Haque, Collinson, and Brook (1965)</td>
<td>England</td>
<td>0.164</td>
<td>0.16 - 0.31</td>
<td>20 measurements, adequate ventilation</td>
</tr>
<tr>
<td></td>
<td></td>
<td>0.35 - 0.40</td>
<td>0.2 - 0.7</td>
<td>20 measurements, inadequate ventilation</td>
</tr>
<tr>
<td>Gemesi, Szy, and Toth (1972)</td>
<td>Hungary</td>
<td>3.1</td>
<td>0.2 - 23</td>
<td>1182 apartments, weighted average of all data</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1.6</td>
<td>0.2 - 6.3</td>
<td>409 brick apartments</td>
</tr>
<tr>
<td></td>
<td></td>
<td>4.6</td>
<td>0.2 - 23</td>
<td>356 slag block apartments</td>
</tr>
<tr>
<td>Parthasarthy, Marshall and Burkinshaw (1972)</td>
<td>England</td>
<td>1.3</td>
<td>0.9 - 1.9</td>
<td>Measured RaA in basement lab</td>
</tr>
<tr>
<td>Toth (1972)</td>
<td>Hungary</td>
<td>3.05 ± 3.66</td>
<td>—</td>
<td>RaA concentration in 841 unventilated apartments</td>
</tr>
<tr>
<td>Jonassen and Hayes (1974)</td>
<td>Denmark</td>
<td>4.1</td>
<td>2 - 8</td>
<td>60 measurements of RaA in a basement room</td>
</tr>
</tbody>
</table>
the mean concentration of 2.5 pCi/L obtained from the analysis given in Fig. 1. The data obtained in Hungary (Gemési, Szy, and Toth, 1972) for apartments constructed with slag blocks yielded a mean concentration of 4.6 pCi/L. However, these apartments present a problem similar to that faced on this continent in using mill tailings for backfill around dwellings. That is, the slag blocks contribute significantly to the radiation environment in the dwellings. For this reason, the use of slag blocks for the construction of apartments was discontinued.

2.3 RADON PROGENY CONCENTRATIONS IN DWELLINGS

Measurements of radon progeny concentrations on the main floor of residential structures in the United States and Canada are presented in Table 4. The data are also presented in the form of a lognormal distribution in Fig. 3. The mean radon progeny concentration is 0.007 WL with a geometric standard deviation of 3.3.

Radon progeny concentrations in basements located in the U. S. and Canada are summarized in Table 5. As before, the data were pooled and plotted in the form of a lognormal distribution (Fig. 4). The data reported by McGregor (McGregor et al., 1980) were not included because approximately 80% of the data were grouped in the range 0.005 WL group which prevented adequate comparison to other data. From these data, the mean radon progeny concentration in basements is 0.014 WL and the geometric standard deviation is 3.5.

In contrast to the data on radon, there have been a number of measurements of WL in dwellings built on reclaimed or altered land. These data are summarized in Table 6 and the distribution is shown in
<table>
<thead>
<tr>
<th>Reference</th>
<th>Country, state</th>
<th>Mean (WL)</th>
<th>Range (WL)</th>
<th>Comments</th>
</tr>
</thead>
<tbody>
<tr>
<td>Keith Consulting (1978)</td>
<td>Canada</td>
<td>0.02 ± 0.04</td>
<td>0.0003 - 0.22</td>
<td>223 measurements at Uranium City, Saskatchewan</td>
</tr>
<tr>
<td>George and Breslin (1978)</td>
<td>New York, New Jersey</td>
<td>0.005 ± 0.003</td>
<td>0.001 - 0.029</td>
<td>19 measurements in selected homes</td>
</tr>
<tr>
<td>Dillworth, Secord, Meagher, and Associates (1978)</td>
<td>Canada</td>
<td>0.03 ± 0.04</td>
<td>0.004 - 0.17</td>
<td>19 measurements in Elliot Lake, Ontario</td>
</tr>
<tr>
<td>Roessler, Wethington, and Bolch (1978)</td>
<td>Florida</td>
<td>0.005 ± 0.008</td>
<td>0.001 - 0.032</td>
<td>15 measurements in selected homes</td>
</tr>
<tr>
<td>Lowder, George, Gogolak and Blay (1971)</td>
<td>Tennessee, Florida</td>
<td>0.012 ± 0.019</td>
<td>0.000014 - 0.14</td>
<td>49 measurements in selected locations</td>
</tr>
<tr>
<td>Eadie, Kaufman, Markley, and Williams (1976)</td>
<td>New Mexico</td>
<td>0.005 ± 0.003</td>
<td>0.0007 - 0.011</td>
<td>17 measurements in Grants Mineral Belt, New Mexico</td>
</tr>
<tr>
<td>U. S. Environmental Protection Agency (1975)</td>
<td>Florida</td>
<td>0.008 ± 0.009</td>
<td>0.0002 - 0.025</td>
<td>9 measurements in two counties, original measurements</td>
</tr>
<tr>
<td>Fitzgerald, Guimond, and Shaw (1976)</td>
<td>Florida</td>
<td>0.012 ± 0.017</td>
<td>0.0002 - 0.027</td>
<td>9 measurements in two counties, new data</td>
</tr>
<tr>
<td>State of Florida (1978)</td>
<td>Florida</td>
<td>0.005 ± 0.005</td>
<td>0.001 - 0.022</td>
<td>34 measurements in Polk and Hillsborough Counties</td>
</tr>
<tr>
<td>Yeates, Goldin, and Moeller (1972)</td>
<td>Massachusetts</td>
<td>0.00058 ± 0.00057</td>
<td>0.0001 - 0.0018</td>
<td>10 single and multiple family dwellings</td>
</tr>
</tbody>
</table>
Fig. 3. Radon progeny concentrations in residential structures in the United States and Canada — data from main floor only.
Table 5. Radon progeny concentrations in residential dwellings, basements only — United States and Canada data

<table>
<thead>
<tr>
<th>Reference</th>
<th>Country, state</th>
<th>Mean (WL)</th>
<th>Range (WL)</th>
<th>Comments</th>
</tr>
</thead>
<tbody>
<tr>
<td>Keith Consulting (1978)</td>
<td>Canada</td>
<td>0.029 ± 0.042</td>
<td>0.0001 - 0.261</td>
<td>251 measurements at Uranium City, Saskatchewan</td>
</tr>
<tr>
<td>George and Breslin (1978)</td>
<td>New York, New Jersey</td>
<td>0.010 ± 0.007</td>
<td>0.0009 - 0.067</td>
<td>19 measurements in selected homes</td>
</tr>
<tr>
<td>George, Breslin and Guggenheim (1976)</td>
<td>New York, New Jersey</td>
<td>0.008 ± 0.004</td>
<td>0.006 - 0.015</td>
<td>6 measurements during a test of cumulative environmental radon monitoring</td>
</tr>
<tr>
<td>George (1972)</td>
<td>New York</td>
<td>0.023 ± 0.004</td>
<td>0.016 - 0.030</td>
<td>11 measurements in basement of New York City office building</td>
</tr>
<tr>
<td>Lowder, George, Gogolak and Glay (1971)</td>
<td>Tennessee, Florida</td>
<td>0.008 ± 0.008</td>
<td>0.001 - 0.026</td>
<td>8 measurements in selected locations</td>
</tr>
<tr>
<td>Spitz and Wrenn (1974)</td>
<td>Colorado</td>
<td>0.131 ± 0.018</td>
<td>0.113 - 0.157</td>
<td>6 measurements in Grand Junction, Colorado</td>
</tr>
<tr>
<td>McGregor et al.* (1980)</td>
<td>Canada</td>
<td>0.0009 - 0.0036</td>
<td></td>
<td>9999 measurements in 13 Canadian cities</td>
</tr>
</tbody>
</table>

From Figure 4: Mean concentration - 0.014 WL; geometric standard deviation - 3.5.

*Not included in Figure 4 (see text for discussion).
Fig. 4. Radon progeny concentrations in residential structures in the United States and Canada — data from basements only.
Table 6. Radon progeny concentrations in residential structures, dwellings constructed on altered or reclaimed land — United States and Canada data

<table>
<thead>
<tr>
<th>Reference</th>
<th>Country, state</th>
<th>Mean (WL)</th>
<th>Range (WL)</th>
<th>Comments</th>
</tr>
</thead>
<tbody>
<tr>
<td>Roessler, Wethington, and Bolch (1978)</td>
<td>Florida</td>
<td>0.027 ± 0.04</td>
<td>0.002 - 0.140</td>
<td>20 measurements in selected homes</td>
</tr>
<tr>
<td>Eadie, Kaufman, Markley, and Williams (1976)</td>
<td>New Mexico</td>
<td>0.014 ± 0.008</td>
<td>0.0013 - 0.037</td>
<td>29 measurements in Grants Mineral Belt, New Mexico</td>
</tr>
<tr>
<td>U. S. Environmental Protection Agency (1975)</td>
<td>Florida</td>
<td>0.054 ± 0.064</td>
<td>0.003 - 0.20</td>
<td>19 measurements in 2 counties, original measurements</td>
</tr>
<tr>
<td>Fitzgerald, Guimond, and Shaw (1976)</td>
<td>Florida</td>
<td>0.035 ± 0.034</td>
<td>0.002 - 0.11</td>
<td>12 measurements in 2 counties, new data</td>
</tr>
<tr>
<td>State of Florida (1978)</td>
<td>Florida</td>
<td>0.013 ± 0.012</td>
<td>0.002 - 0.069</td>
<td>97 measurements in Polk and Hillsborough Counties</td>
</tr>
</tbody>
</table>

From Figure 5: Mean concentration = 0.011 WL; geometric standard deviation = 2.8.
Fig. 5. The mean concentration, determined from the graph, is 0.011 WL with a geometric standard deviation of 2.8.

3. OTHER CONSIDERATIONS OF RADON EXPOSURE CONDITIONS

The ubiquitous presence of radium-226 in the surface of the earth provides a continuous source of radon and radon progeny to which the human population is exposed. However, the environmental conditions prevailing during this continuous human exposure are by no means constant. It is clear that the exposure conditions are quite variable since only a small percentage of the population remains in one particular location 100% of the time. In addition, the environmental factors associated with the exposure have a significant effect on the exposure situation.

The major environmental factors which relate to radon-222 daughter dosimetry include: (1) the radon-222 concentration, (2) the degree of equilibrium* between radon-222 and its daughters, and (3) the fraction of radon-222 daughters existing as free ions. The magnitude of one or more of these factors is affected by many variables. These variables will be examined below as they have been reported for nonmining situations.

*In this paper, the state of equilibrium (disequilibrium) will refer to the daughter concentrations relative to the radon-222 concentration. The ratio will be expressed as Rn:RaA:RaD:RaC.
Fig. 5. Radon progeny concentrations in residential structures in the United States and Canada — data from dwellings built on altered or reclaimed land.
3.1 RADON CONCENTRATIONS

It is well known that radon-222 contributes only a small fraction of the total absorbed dose caused by its daughters (Hultqvist, 1956). Thus, it is common to consider radon-222 to be an isotope generator; the source of the radon daughter activity. For this reason, it is usually not sufficient to measure only the radon-222 concentration and apply a single absorbed dose conversion factor. It is imperative that the true average concentration be estimated as accurately as possible. Haque, Collinson, and Brook (1965) reviewed sampling techniques and results and recommended that the sampling time should be conducted over a total time of about 3 to 4 weeks. Furthermore, these authors advise the selection of a suitable time of the day for sampling which could be selected on the basis of preliminary measurements. Average concentration values obtained by the methods outlined by Haque and his colleagues in their opinion should not deviate from the true mean by more than 20 to 30% (Haque, Collinson, and Brook, 1955).

Daily variations in the radon-222 concentration in single-family structures built on tailings fill have been measured by Spitz and Wrenn (1974). These investigators, using a continuous radon monitor, noted a daily variation in the radon-222 concentration of 2 orders of magnitude in a vacant house, whereas the variation in an occupied dwelling was 3 orders of magnitude. The radon concentration in a previously sealed house decreased substantially when the house was opened. Although the authors give no suggested numerical values, they recommend that an occupancy factor be used to approximate the large variation in the radon-222 concentrations encountered.
Hultqvist (1956) also surveyed radon-222 concentrations within dwellings. However, daily variations in the concentrations reported spanned only 1 order of magnitude. Furthermore, the times at which maximum and minimum concentrations occurred were approximately the same for the three apartments surveyed. No temporal relationship of this type was observed among the six residences surveyed by Spitz and Wrenn (1974).

The average radon concentration values obtained by Hultqvist were in the range of 1 pCi/L. In contrast, those obtained by Spitz and Wrenn ranged from tens of pCi/L to almost 100 pCi/L. It appears reasonable, based on the results of these two investigations, to expect a daily range of radon measurements to be from one-third to three times the average. In addition, Hultqvist (1956) found weekly and seasonal variations in the daily average radon concentrations. These variations ranged between one-half to twice the average. All of the variations discussed above are variations within a given residence. Spitz and Wrenn (1974) also found room to room variations within the same residence.

A survey of radon-222 concentrations in Hungarian residential structures by Gemesi, Szy, and Toth (1972) revealed that concentrations of this noble gas varied from structure to structure, being quite dependent on the materials of construction. For example, in buildings constructed of sun-dried unburnt clay, the average radon-222 concentration was 1.70 pCi/L with minimum and maximum concentrations of 0.2 and 5.8 pCi/L, respectively. For residences constructed of slagblocks, the average concentration was 4.60 pCi/L with minimum and maximum concentrations of 0.2 and 22.5 pCi/L, respectively. A total of 1182 measurements were made in this survey with a weighted average concentration of 3.05 pCi/L.
Hultqvist (1956) also found high variability among different structures. Data of other investigators, assembled by Hultqvist, also showed considerable variability. Variations in the radon concentration among residences observed by Spitz and Wrenn (1974) prompted these authors to caution that each structure should be considered as a separate entity. Haque, Collinson, and Brook (1965) observed that radon-222 concentrations reached maximum values in rooms which had been sealed for eight days. However, of the three sealed rooms investigated, the equilibrium, or ultimate concentration was a function of the exhalation rate of the wall material.

The investigations reviewed above are not the only surveys of radon-222 concentrations in structures which have been reported. According to Hultqvist (1956), measurements of this type have been made by other investigators since about 1905. Measurements made by other investigators generally fall into two diverse categories. In some cases, the data exhibit a wide variability and, in other cases, the data show less deviation from the average concentration. However, the sources reviewed indicate clearly that a single grab sample obtained at a location may lie anywhere on the distribution of concentration values. Thus, it seems appropriate to assume that the true average radon-222 concentration will lie between one-third and three times the grab sample concentration. This relationship may be better defined by a thorough statistical analysis of the distribution of reported concentrations.
The available data also indicate that great care should be used in the extrapolation of data obtained at one location to the expected concentration at another location. Such extrapolations may be in error by as much as a factor of 7.

3.2 RADON PROGENY EQUILIBRIUM

The radon daughters are the radionuclides which actually deposit their energy in lung tissue. In a hypothetical situation of continuous exposure to radon and radon daughters in one location, it is easy to formulate the extreme exposure conditions. The maximum exposure results from the inhalation and retention of the material until every atom of polonium-214 generated by the airborne radon-222 has decayed. The minimum exposure, of course, would result if all the daughter atoms were collected or swept away before inhalation. Obviously, the extreme maximum is physically impossible for a human with an air intake rate of 15 L/min. Likewise, the extreme minimum is physically impossible since it would require an infinite supply of radon-free dilution air or a respirator with an absolute filter.

Tsivoglou, Ayer, and Holoday (1953) presented calculations which showed that the inhalation hazard associated with radon-222 and daughters was dependent strongly on the state of equilibrium attained. For example, the absorbed dose associated with very low equilibrium (1:0.1:0.5:0.0) is only 4% of the absorbed dose at complete equilibrium (1:1:1:1). More recently, Holaday and Jones (1973) have shown that changes in the ratios among daughters have little effect on the magnitude of the inhalation hazard. However, these authors also demonstrated that changes in the
radon to daughter ratios have a marked effect on the inhalation hazard. Thus, estimates of the absorbed dose in the human lung based on a measurement of radon-222 concentration requires a careful evaluation of the state of equilibrium which exists between radon-222 and its daughters.

If the airborne radon daughters were subject only to the laws of radioactive decay, equilibrium would develop in a manner described by Evans (1969). However, many parameters affect the degree of equilibrium which exists in practical exposure situations. Tsivoglou, Ayer, and Holoday (1953) recognized the influence of ventilation on the degree of equilibrium attained. Hultqvist (1956) presented a series of simultaneous equations which expressed the concentration of each radon daughter as a function of the ventilation rate. Haque and Collinson (1967) whose work was the cornerstone of that of Johnson, Hardin, and Nelson (1975) used a modification of Hultqvist's equations to calculate the radon daughter concentrations (no radon daughter measurements were made by Haque and Collinson).

The alpha particle emitted from the radon-222 nucleus actually strips some of the orbital electrons from the decaying atom. Thus, the polonium-218 daughter is born as a free ion and subsequently is subject to electric and magnetic forces which may exist. These ions can be neutralized or absorbed onto negatively charged surfaces, such as airborne particles or surfaces in a room (walls, furniture, etc.) which become a sink for radon daughters. In the absence of ventilation, the airborne radon-222 will attain secular equilibrium with its daughters. However, a fraction of the daughters will be absorbed on surfaces of the room. Wrenn, Rosen, and Van Pelt (1969) have presented a theoretical approach
for the estimation of daughter deposition under quiescent conditions. Jacobi (1972a) also has calculated the simultaneous effects of ventilation and deposition on the state of daughter equilibrium.

Even though the effects of ventilation and deposition on radon daughter equilibria are understood under constant conditions, these parameters are also subject to variations which tend to further complicate the true situation. For example, Handley and Barton (1973) recommend a range of 0.5 to 1.5 air changes per hour be used in the evaluation of absorbed dose from radioactive contaminants in natural gas. These authors conclude: "Available data are insufficient to assign a single value for an average or characteristic occupied house." The effect of this range of ventilation rate may be estimated using the simplified equations of Haque and Collinson (1967). These calculated daughter ratios, presented in Table 7, reflect that the RaB and RaC ratios to radon are proportional to the inverse square and the inverse cube of the ventilation rate, respectively.

However, as mentioned previously, ventilation is not the only process which influences airborne radon-222 daughters concentrations. Toth (1972) surveyed airborne radon daughters in 841 residences in Hungary. In 92% of these investigations, the surveyed rooms had been sealed for at least 8 hours prior to sampling. Radon-222 and polonium-218 concentrations were measured simultaneously in 98 of the sealed rooms. The mean radon-222 concentration was equivalent statistically to the mean polonium-218 concentration. Extrapolating this relationship to the remainder of the 841 measurements, the average of the daughter ratios to radon was 1:1:0.87:0.82. Evans (1969) showed that, for all practical purposes,
Table 7. Relation between ventilation rate and radon daughter equilibrium

<table>
<thead>
<tr>
<th>Air exchange rate (h⁻¹)</th>
<th>Rn:RaA:RaB:RaC Ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.5</td>
<td>1:0.96:0.73:0.59</td>
</tr>
<tr>
<td>1.0</td>
<td>1:0.93:0.57:0.39</td>
</tr>
<tr>
<td>1.5</td>
<td>1:0.90:0.46:0.27</td>
</tr>
</tbody>
</table>
radon-222 will obtain full secular equilibrium with its daughters after a period of three hours. The data obtained by Toth (1972) provide some qualitative experimental verification of the deposition theory of Wrenn, Rosen, and Van Pelt (1969), and Jacobi (1972a).

In addition to removing airborne radon daughters from the room under consideration, ventilation also increases deposition on room surfaces (Wrenn, Rosen, and Van Pelt, 1969; Jacobi, 1972a). Jonassen (1975) indicated that the enhancement of deposition by ventilation may be far more effective in removing airborne radioactivity than that attributable solely to ventilation. Specifically, the elimination constants (lambda) for deposition on a basement wall are about an order of magnitude higher than the elimination constant due to ventilation. However, Jonassen cautions that the relationship may hold only for the particular set of measurements obtained in his research.

Parthasarathy, Marshall, and Burkinshaw (1972) have reported that synthetic substances such as nylon, polyvinyl chloride, "Fablon," and polyethylene strongly absorb radon daughters due to their high surface static charge. The deposition of daughters was markedly inhibited by applying an antistatic spray to the surface of the polyethylene sheet. The authors note that untreated surfaces of this type material continually carry a residual charge. Since it has been demonstrated that 50% of particles carrying radon daughter products are charged positively and 25% negatively (Sollieux, 1970), these authors recommend that the surfaces on which deposition can occur should be electrically neutral.
Yeates, Goldin, and Moeller (1972) reported radon daughter and ventilation rate measurements in single family dwellings, multifamily dwellings, and office buildings. Concentrations of radon-222 were not measured in this study. The data obtained should not be used to establish the relationships between radon daughter concentrations and ventilation rate because the very low concentrations encountered approached the sensitivity of the measuring techniques employed. Schiager and Olson (1971) measured radon-222, radon progeny as working levels, and ventilation rates in experimental structures, however, because the equipment used to determine progeny concentrations subsequently was found to be faulty, these data cannot be used.

3.3 FREE ION FRACTION

The fraction of radon daughters existing as free ions in air has an important influence on the absorbed dose to the lung due to the inhalation of radioactivity. This fraction, called the unattached fraction $f$, is of prime importance in dosimetry calculations (particularly RaA) involving radon daughters (Holaday and Jones, 1973; Walsh, 1970). Haque and Collinson (1967) used the aerosol data obtained by Mohnen and Stierstadt (1963) and calculated a value of 0.35 for the unattached fraction. They obtained this unattached fraction from the formula

$$f = \frac{\lambda_0}{\lambda_0 + \lambda_p}$$
where $\lambda_o$ is the radiological decay constant and $\lambda_p$ is the attachment constant. The equation for $\lambda_p$ given by Lassen, Rau, and Weicksel (1960, 1961) based on neutral atom diffusion was used in the calculation.

Mohnen (1968) in a comprehensive analytical study measured the attachment coefficient as a function of particle size. Using the appropriate attachment coefficient from this study and the data used by Haque and Collinson (1967), the value of $f$ is 0.08. If the aerosol data obtained by George (1972) is used, with the appropriate data from Mohnen (1968), $f$ has a value of 0.06.

George (1972) measured the uncombined fraction of RaA at both the basement and fifth floor locations in an office building. Although the mean RaA concentrations were significantly different between the two locations, the mean uncombined fractions were not significantly different. A value of 0.06 is obtained for $f$ from the average of the measured data. This is the same value estimated by using the aerosol data of George (1972) and the coefficient reported by Mohnen (1968). The true mean of the $f$ values measured by George lies between 0.05 and 0.07 at the 95% confidence level. Applying the technique of Haque and Collinson (1967) to the data of George (1972), the $f$ value obtained is 0.09. Thus, in terms of the data presented by George (1972), it appears that Mohnen's work (1968), based on considerations of neutral atom and ion interactions, is far superior to the formula of Haque and Collinson (1967) which was based solely on neutral atom diffusion.
3.4 PARTICULATE CONCENTRATION

A factor which has not been discussed in this review is the particulate content of the atmosphere. The particulate content is characterized by the concentration of particles and the particle size distribution. These considerations are important in dosimetry because particle properties influence strongly the site and efficiency of energy deposition in the human respiratory system. Furthermore, radon daughters which are attached to airborne particles are less likely to deposit on electrically charged room surfaces (Wrenn, Rosen, and Van Pelt, 1969; Jacobi, 1972a). Thus, it is clear that the concentration of airborne particles also influences the concentration of airborne radon daughters.

A number of equations have been developed to calculate the rate at which daughters are absorbed onto particles (e.g., see 21, 22, and 24). Basically, all equations relate the attachment rate ($\lambda_p$) to the surface area and concentration of particles.

Jacobi (1972a,b) has derived expressions relating the state of equilibrium of radon daughters to particle concentration, ventilation rate, and wall deposition. To estimate the state of equilibrium existing in a hypothetical home, estimates of wall deposition and particle concentration must be made. For his calculations, Jacobi (1972a,b) assumed a surface to volume ratio of 2 m$^{-1}$ which he takes as typical of mining situations. This assumption yields a wall deposition rate ($\lambda_d$) of 0.7 h$^{-1}$. For air exchange rates in the range of 0.5 to 1 per hour, it must be assumed that $\lambda_d = \lambda_v$; a conservation assumption based on the work of Jonassen (1975).
In a home of the size assumed by Johnson, Hardin, and Nelson (1975), a surface to volume ratio of 2 m$^{-1}$ is within the range of physical possibility. In a larger home, the ratio may be smaller, but room to room variations in other variables will introduce greater sources of error.

The particle size distribution in the postulated constant exposure situation should be evaluated at this point. Johnson, Hardin, and Nelson (1975) used the assumptions of Haque and Collinson (1967) regarding particle distribution. Haque and Collinson (1967), in turn, used particle size distribution and total particle content data obtained during the investigations of Mohnen and Stierstadt (1963). Mohnen, in a later published work (1968), reported that his experiments were performed deliberately between the hours of midnight and 5 a.m. to decrease the dependence of the particle spectrum with time. He states: "Because of the deliberately chosen small resolution of the spectrograph, the size spectrum has only a limited information value. The total concentration $Z = 3.2 \times 10^4$ particles/cm$^3$ is only valid for the period between midnight and 5 a.m. His data (Fig. 22) indicates that the total particle concentration at other times of the day is usually much greater than the $3 \times 10^4$ cm$^{-3}$ which has been quoted so extensively. A reasonable average particle concentration interpolated from his data would be about $5 \times 10^4$ cm$^{-3}$. However, the data presented were obtained over a period of less than 24 hours and may not represent a typical day.

George (1972) made aerosol measurements in the basement and fifth floor of a New York City office building. His measurements included average particle size, particle distribution (expressed as the geometric
standard deviation, $\sigma_g$, and particle concentration. The pooled data for the basement and fifth floor measurement, which were not significantly different, yielded a mean particle size of 0.14 μm activity median diameter (AMD), a geometric standard deviation, $\sigma_g$, of 3.0, and a particle concentration, $c$, of 46,000 cm$^{-3}$.

The chemical composition of particles in urban air may range from organic matter (specific gravity <1), water (s.g. = 1), soot (variable s.g.), and dust (s.g. ~2.7). A reasonable average of the specific gravities is about 2. Thus, the AMD found by George (1972) corresponds to a sphere of radius 0.05 μm. Assuming a spherical shape and a density of 2 gm/cm$^3$, the particle spectrum measured by George (1972) represents a particulate content of about 40 μgm/m$^3$. For purposes of comparison, the national average USPHS Air Quality Data (1965) showed a geometric mean of 90 μgm/m$^3$ for urban and 28 μgm/m$^3$ for nonurban reporting stations. However, indoor aerosols cannot be compared directly to outdoor aerosols as the composition of the two differs drastically. The particle concentration reported by George (1972) corresponds roughly to that interpolated from the data of Mohnen (1968). In addition, residential particle concentrations will vary depending on the smoking habits of the residents, the degree of household cleanliness, and the region of the country in which the residence is located.

The particle size obtained by George (1972) corresponds approximately to the mid-range of particle sizes measured for tobacco smoke and dust. In addition, this particle size compares favorably with the value of 0.2 μm found by Mercer and Stowe (Johnson, Hardin, and Nelson, 1975). Combining the data of George (1972) for particle distribution, previously
discussed ventilation rates, and the wall deposition rate from the method of Jacobi (1972a), the equilibrium ratio for radon daughters to radon is ^1:0.9:0.4:0.2. The average ratios obtained from the data reported by George (1972) are: basement - 1:0.86:0.41:0.22; fifth floor - 1:0.92:0.38:0.23. These experimental data, although representing only one location, are in agreement with estimates obtained by using the stated assumptions and the method of Jacobi (1972a). However, the equilibrium ratios obtained from the data reported by George (1972) vary widely from day-to-day over a two-month period. Thus, it must be recognized that the average condition, represented by the equilibrium ratio 1:0.9:0.4:0.2, should be used with extreme caution until additional data are available.

Although there is a dearth of experimental data related to domestic radon and radon progeny exposure conditions, it is possible on the basis of the previous review to assign some probable values to the various parameters. These data are given in Table 8 and, since the data are estimates, all values have been rounded to one significant figure. An estimate of the "worst case" exposure is included in the table although it is doubtful that the rigorous conditions necessary for such an exposure could be maintained for periods of time representing a continuous exposure.
Table 8. Summary of radon exposure conditions for 1 pCi/L of radon-222

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Probable mean</th>
<th>Probable range</th>
<th>Worst case</th>
</tr>
</thead>
<tbody>
<tr>
<td>RaA (pCi/L)</td>
<td>0.9</td>
<td>0.6-1.0</td>
<td>1.0</td>
</tr>
<tr>
<td>RaB (pCi/L)</td>
<td>0.4</td>
<td>0.2-0.8</td>
<td>0.9</td>
</tr>
<tr>
<td>RaC (pCi/L)</td>
<td>0.2</td>
<td>0.1-0.6</td>
<td>0.8</td>
</tr>
<tr>
<td>Unattached fraction for RaA, f</td>
<td>0.06</td>
<td>0.04-0.1</td>
<td>0.2</td>
</tr>
<tr>
<td>Aerosol diameter, AMD (µm)</td>
<td>0.14</td>
<td>0.09-0.2</td>
<td>0.07</td>
</tr>
<tr>
<td>Aerosol distribution, $\sigma_g$</td>
<td>3</td>
<td>2-4</td>
<td>~1</td>
</tr>
<tr>
<td>Aerosol concentration (cm$^{-3}$)</td>
<td>40,000</td>
<td>20,000-80,000</td>
<td>10,000</td>
</tr>
</tbody>
</table>
4. DISCUSSION

The most noticeable feature of this study is that both radon and radon daughter concentrations in structures built on a particular land type may vary over several orders of magnitude. The radon concentrations and geometric standard deviations in structures built on unaltered lands were found to be 2.5 pCi/L ± $\sigma_g = 4.8$ and 7.1 pCi/L ± $\sigma_g = 3.5$ for first floor and basement measurements, respectively. In structures built on altered lands, the values have been reported to be 4.0 ± 6.29 pCi/L (Roessler, Wethington and Bolch, 1978). The radon progeny concentrations and geometric standard deviations in structures built on unaltered lands were found to be 0.007 WL ± $\sigma_g = 3.3$ and 0.014 WL ± $\sigma_g = 3.5$ for first floor and basement measurements, respectively. A summary of radon progeny measurements in structures built on altered lands yielded a value of 0.011 WL ± $\sigma_g = 2.8$.

These statistical results indicate that the lognormal distribution is adequate for representing the indoor radon and radon daughter concentration data. It should be carefully noted that geometric standard deviations in the range obtained above (2.8–4.8) indicate a serious sampling problem. Any evaluation of radon and radon progeny in structures associated with a particular region or land type must include a sufficient number of measurements to adequately define the distribution. Further, sampling needs are complicated by the temporal variations, both diurnal and seasonal, in radon concentration in structures of one order of magnitude or more as discussed in Section 3.1.
Radon progeny are those radionuclides which contribute to inhalation dose and because the inhalation hazard is strongly dependent on the degree of equilibrium among radon and radon progeny, sampling should address these considerations as well as factors discussed in Sections 3.2, 3.3, and 3.4. In order to adequately describe the radon and radon progeny distribution in structures built on a particular land type or in a particular area, it appears necessary to design a sampling program which will allow evaluation of the temporal and physical factors which will affect radon and radon progeny concentrations.
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APPENDIX

Surgeon General of The United States:

Recommendations for Remedial Action in Dwellings Constructed on or with Uranium Mill Tailings
<table>
<thead>
<tr>
<th>External gamma Radiation level</th>
<th>Indoor radon daughter concentration</th>
<th>Recommendation</th>
</tr>
</thead>
<tbody>
<tr>
<td>Greater than 0.1 mR/hr</td>
<td>Greater than 0.05 WL</td>
<td>Remedial action indicated</td>
</tr>
<tr>
<td>From 0.05 to 0.1 mR/hr</td>
<td>From 0.01 to 0.05 WL</td>
<td>Remedial action may be suggested</td>
</tr>
<tr>
<td>Less than 0.05 mR/hr</td>
<td>Less than 0.01 WL</td>
<td>No remedial action indicated</td>
</tr>
</tbody>
</table>

*For use within dwellings or structures constructed on or with uranium mill tailings — levels given are assumed to be above background for the area.*
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