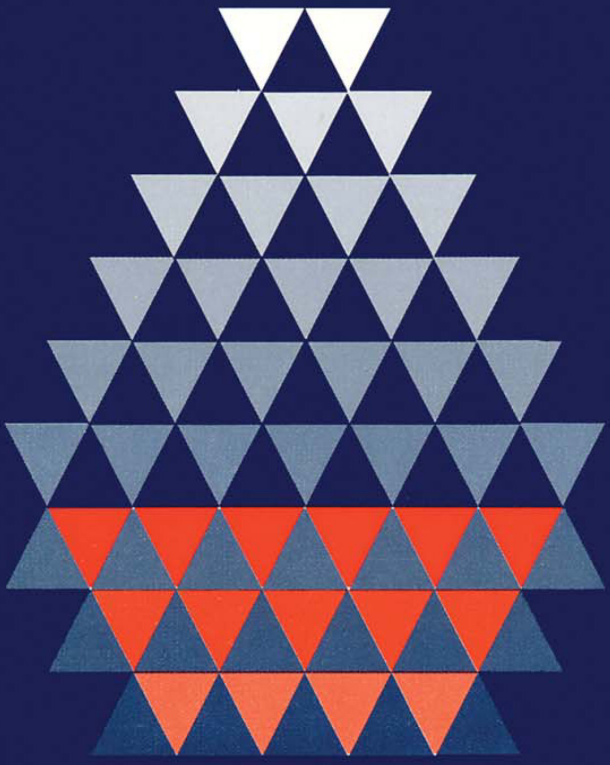

# Radon, Radium and Uranium in Drinking Water

Edited by

C. Richard Cothern

Paul A. Rebers

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Dr. Rebers earned a BS and MS in Chemical Engineering and a PhD in Biochemistry from the University of Minnesota. He has published over 60 papers on the structure of polysaccharides and on the immunochemistry of animal disease. Dr. Rebers is a member of the Graduate Faculty at Iowa State University.

# Preface

This single volume covers all aspects concerning radionuclides in drinking water. The primary focus is on the naturally occurring radionuclides, primarily radon, radium, and uranium. The specific topic areas covered include occurrence, mechanisms resulting in human exposure, analytical chemistry, health effects, quantitative risk assessment, treatment technology for the removal of radionuclides, waste disposal, economics, policy, and some aspects of enforcement.

A wide range of people in the United States are exposed to radionuclides in drinking water. As this volume points out, perhaps the group most at risk are those with private wells. The health consequences of radionuclides in drinking water are nationwide for both public and private water supplies, and all need to understand the risks involved.

The U.S. Environmental Protection Agency in the process of developing regulations for radionuclides in drinking water to update the interim regulations promulgated in 1976. The earlier regulations covered radium, and the new ones will include radon and uranium. This broader regulatory coverage will involve many new communities that were previously in compliance with the interim regulations. This volume is written for the regional, state, and local health officials who need to understand the background information involved in complying with the regulations for radionuclides in drinking water. Those involved in all levels of providing public drinking water will find details in this volume concerning exposure to radionuclides, the resulting estimated health effects, treatment options, and discussions of the economics and policy consequences of compliance.

The reader may wish to find more details on the subjects in this volume. The following represent a suggested list of supplemental reading: the May 1985 issue of the journal *Health Physics* entitled "Special Issue on Radioactivity in Drinking Water"; *Federal Register*, Tuesday, September 30, 1986, pp. 34836–62, "Advanced Notice of Proposed Rulemaking for Radionuclides in Drinking Water"; a supplemental textbook *Environmental Radon*, published in 1987 by Plenum Publishing Corp., 233 Spring Street, New York, NY, pp. 10013-1578; and the book, *Health Effects of Radon and Other Alpha Emitters*, BEIR IV, published in 1988 by the National Academy Press, Washington, DC.

Two basic systems of units are used in this book: the historical and the SI or international system (a complete discussion of these units can be found in the text *Environmental Radon*, mentioned above). The historical units include curies (Ci) for activity, rem for dose equivalent, and working level for radon

exposure units. The corresponding SI units are becquerels (Bq), sieverts (Sv), and joules per cubic meter. Conversions that may be useful here are 1 Bq = 27 pCi (where p or pico means $10^{-12}$) and 1 mSv = 100 mrem.

The thoughts and ideas discussed in this book are those of each individual author and do not necessarily reflect the policies of the U.S. Environmental Protection Agency.

C. Richard Cothern and Paul Rebers

# Contents

# CHAPTER 1

# Scientific Background for the Development of Regulations for Radionuclides in Drinking Water

**Paul Milvy and C. Richard Cothern**

## INTRODUCTION

Nearly all of the radionuclides occurring in drinking water supplies are naturally occurring. The radionuclides in drinking water will vary somewhat according to the source of the water. Under the Safe Drinking Water Act, *public drinking water supplies* are defined as those that serve 25 or more people or have 15 or more service connections. The water for 80% of the nation's 60,000 public water supplies comes from ground water sources. Nearly 70% of the population is served by the public drinking water supplies that use surface water.

The radionuclides in drinking water are members of three natural radioactive series. These are the uranium series, the thorium series, and the actinium series. The specific elements of concern are radium (Ra), radon (Rn), and uranium (U). The isotopes in the uranium series that may pose a health risk because of their presence in drinking water are $^{238}U$, $^{234}U$, $^{226}Ra$, and $^{222}Rn$. Radium-228, which occurs in the thorium series, is also found in drinking water. Very few of the other isotopes in these series have been detected in drinking water. Recently, however, polonium-210 has been detected in the drinking water of east central Florida.[1]

In 1976, interim drinking water standards were enacted into law that limited concentrations in public water supplies of gross alpha particle activity, and the combined concentration of $^{226}Ra$ and $^{228}Ra$. The U.S. Environmental Protection Agency is presently developing final standards for uranium, radium, and radon in drinking water. This chapter discusses the occurrence, health effects, and risk assessments of these naturally occurring radionuclides in drinking water, considering them in the context of the U.S. Environmental Protection Agency's maximum contaminant level (MCL) selection process.

For additional details concerning radionuclides in drinking water, see the background scientific data discussed in the advanced notice for proposed rulemaking and a general article on radon in drinking water.[2,3] For more details

concerning the general area of environmental radon, the reader is directed to a supplemental textbook.[4]

## OCCURRENCE OF NATURALLY OCCURRING RADIONUCLIDES IN DRINKING WATER

### Radium

The occurrence data for radium in drinking water come from the compliance record provided by the states from surveys conducted as required by the Interim Drinking Water Regulations (also see some recent data discussed in chapter 8). From this data it is estimated that approximately 500 of the approximately 60,000 public drinking water supplies have radium levels that exceed 5 pCi/L, the interim MCL set in 1976.[5] The interim drinking water standard applies to combined $^{226}Ra$ and $^{228}Ra$. Although the occurrence of $^{226}Ra$ and $^{228}Ra$ are independent (they occur in two different radioactive series), in general their nationwide distributions with respect to concentration are somewhat similar. The largest concentration that has been measured in a public drinking water supply is approximately 100 pCi/L; the mean for $^{226}Ra$ is about 0.4 pCi/L, and the mean for $^{228}Ra$ is about 0.7 pCi/L.

### Uranium

Knowledge of the uranium concentrations in public drinking water supplies in the United States has been derived, in part, from information based on a survey conducted during the 1960s and 1970s by the U.S. Geological Survey called the National Uranium Resource Evaluation (NURE). The NURE study segmented the United States into areas of about 10 kilometers on a side. In each of these areas, a groundwater, surface water, and sediment sample were taken. Although the survey was not completed for all areas, approximately 89,000 ground and surface water samples were collected. A more recent survey, designed in 1983 and completed in 1988, is known as the National Inorganics and Radionuclide Survey (NIRS). It involved measurements of the concentrations of uranium and radium-226 and -228 in groundwater sampled at 1000 sites.

Most measurements of uranium are made on the basis of its mass rather than its radioactivity. At secular equilibrium, the ratio of $^{234}U$ to $^{238}U$, since they are in the same radioactive series, is one. However, in many water supplies this ratio is greater than one and can be, in fact, as large as 10 or 20 because of selective recoil of the different isotopes at the rock/water interface. This is related to the mechanisms by which the isotopes, originally present in soil and rock, become soluble and enter the aquifers. Since the half lives of these isotopes are grossly disparate ($2.5 \times 10^5$ vs $4.5 \times 10^9$ years, respectively), if equilibrium is wrongly assumed, converting the mass measurement to radioac-

tivity underestimates the level of uranium present. Nevertheless, a conversion factor that equates one μg of uranium to 0.67 pCi of uranium is commonly used.

The minimum detection concentration for uranium in drinking water is 0.5 pCi/L, and the largest measured level in the NURE study is 600 pCi/L (although a concentration of 6900 pCi/L was indicated by a recent report from a private well west of Denver, Colorado[6]). The Oak Ridge National Laboratory has identified approximately 20,000 of the sites for the NURE study as either existing drinking water supplies or potential drinking water supplies and found that the mean concentration for these supplies equals 1.8 pCi/L.[7]

## Radon

Radon in drinking water is found only in groundwater supplies (the insoluble radon gas quickly degasses in surface water supplies). Smaller water supplies tend to take their water from smaller aquifers, which, in general, have larger granular surface areas and, thus, higher concentrations of radon. Consequently, the levels are higher for supplies providing water to smaller communities. It is expected, based upon this trend as well as additional limiting sampling, that radon occurrences in private wells will be a factor of three to ten times higher than concentrations measured in public drinking water supplies. The minimum detection level for routine analytical measurements for radon in drinking water is in the range of 50 to 100 pCi/L, and the largest value observed in a public water supply is 750,000 pCi/L. The average U.S. groundwater concentration of radon is in the range of 200 to 600 pCi/L.

Waterborne radon enters a house through the water supply and is released into the indoor air when the water is used for showers, bathing, washing clothes, washing dishes, cooking, and flushing toilets. The most important exposure route for radon from drinking water is the inhalation route via the above release mechanisms, and, most likely, less exposure to radon occurs by virtue of the ingestion of water or beverages. It has been estimated that the air concentration of radon released from water is 1/10,000 that of the water's concentration of radon. Thus, if the water concentration of radon is 10,000 pCi/L of water, an average of 1 pCi/L of air is contributed to the indoor air levels. The median U.S. indoor air concentration of radon is about 1 pCi/L of air, of which between 1 and 7% has its origin in the water supply.[8] The outdoor radon levels are on the order of 0.1 pCi/L of air. Thus, a radon concentration of 1000 pCi/L in water would contribute to indoor air an amount roughly equal to the average outdoor concentration.

## Geographical Distributions

Although radionuclides in drinking water are found throughout the United States, certain geographic areas have particularly high levels.[9] The highest levels of radon are observed in the New England states and in the Virginia-

**Table 1. Range of Average Radionuclide Concentrations in Units of pCi/L**

| Radionuclide | All Water Supplies | Groundwater Supplies |
|---|---|---|
| Ra-226 | 0.3—0.8 | 0.4 |
| Ra-228 | 0.4—1.0 | 0.7 |
| U-238 | 0.3—2.0 | 1.9 |
| Rn-222 | 50—300 | 600 |
| Pb-210 | < 0.1 | |
| Po-210 | < 0.1 | |
| Th-230 | < 0.1 | |
| Sr-90 | < 0.2 | |

Maryland region. High levels of radium in drinking water are measured in the Piedmont region of North Carolina, in South Carolina and Georgia, the midwestern area of Iowa, northern Illinois and southern Wisconsin, and the four corners region in the West. The highest levels of uranium are found in the mountainous regions of the western United States. The reasons for these distributions can be found in the geological distribution of the material and the geochemical transport mechanisms by which these radionuclides reach the tap. Based on our understanding of geochemistry, the U.S. Environmental Protection Agency has listed counties in the United States that would be expected to fall in high, medium, and low concentration regions. For instance, counties have been identified that may have uranium concentrations that would be below 1 pCi/L, in the range of 1 to 5 pCi/L, in the range of 5 to 10 pCi/L, and greater than 10 pCi/L.[10]

Table 1 shows the average radionuclide concentrations in the U.S. public drinking water supplies. These values represent ranges of a number of individual average values reported in various studies, not the extreme minimum and maximum of individual measurements that were cited earlier in this chapter.

Tables 2 to 5 present the empirical data of the NIRS on the distribution of radium-226, radium-228, radon, and natural uranium activities in the nation's public groundwater supplies. An analysis of the radium data (Tables 2 and 3) shows that the population-weighted average total radium concentration is about 1.1 pCi/L. The average population-weighted concentration of uranium in ground and surface waters in the United States is approximately 1.3 pCi/L.

**Table 2. Occurrence of Radium-226 by Population and Activity**

| Population | Number of Systems With Indicated Activities (pCi/L) | | | | | | |
|---|---|---|---|---|---|---|---|
| | < 0.18[a] | 0.18–1 | 1–2.5 | 2.5–5 | 5–7.5 | 7.5–10 | 10–12.5 |
| 25–1000 | 492 | 238 | 51 | 10 | 2 | 1 | 1 |
| 1001–5000 | 73 | 57 | 7 | 1 | 2 | 0 | 1 |
| 5001–50,000 | 33 | 15 | 3 | 4 | 1 | 0 | 0 |
| > 50,000 | 12 | 1 | 0 | 0 | 0 | 0 | 0 |
| Totals | 610 | 311 | 61 | 15 | 5 | 1 | 2 |

Note: Population-weighted mean = 0.41 pCi/L, assuming that the concentration below the minimum reporting level = 0.09 pCi/L.
[a]Minimum reporting level.

**Table 3. Occurrence of Radium-228 by Population and Activity.**

| Population | Number of Systems With Indicated Activities (pCi/L) | | | | | | |
|---|---|---|---|---|---|---|---|
| | < 1[a] | 1–2 | 2–3 | 3–4 | 4–5 | 5–6 | > 6 |
| 25–1000 | 687 | 75 | 9 | 2 | 2 | 1 | 4 |
| 1001–5000 | 128 | 8 | 0 | 3 | 1 | 1 | 0 |
| 5001–50,000 | 49 | 5 | 3 | 0 | 1 | 1 | 2 |
| > 50,000 | 1 | 0 | 0 | 0 | 0 | 0 | 0 |
| Totals | 865 | 88 | 12 | 5 | 4 | 3 | 6 |

Note: Population-weighted mean = 0.68 pCi/L, assuming that the concentration below the minimum reporting level = 0.5 pCi/L.
[a]Minimum reporting level.

**Table 4. Occurrence of Natural Uranium by Population and Activity**

| Population | Number of Systems With Indicated Activities (pCi/L) | | | | | |
|---|---|---|---|---|---|---|
| | < 0.1[a] | 0.1–5 | 5–15 | 15–30 | 30–60 | > 60 |
| 25–1000 | 226 | 465 | 61 | 11 | 10 | 4 |
| 1001–5000 | 32 | 86 | 19 | 3 | 0 | 0 |
| 5001–50,000 | 13 | 43 | 0 | 0 | 0 | 0 |
| >50,000 | 1 | 2 | 0 | 0 | 0 | 0 |
| Totals | 272 | 596 | 80 | 14 | 10 | 4 |

Note: Population-weighted mean = 1.9 pCi/L, assuming that the concentration below the minimum reporting level = 0.5 pCi/L.
[a]Minimum reporting level.

Table 4 shows the distribution of uranium in groundwater supplies. The distribution of radon in drinking groundwater supplies is shown in Table 5 and, as indicated, covers many orders of magnitude. The population-weighted average concentration of radon in drinking water from groundwater sources in the United States is several hundred pCi/L (the median is about 200 pCi/L).

**Table 5. Occurrence of Radon-222 by Population and Activity**

| Population | Number of Systems With Indicated Activities (pCi/L) | | | | | |
|---|---|---|---|---|---|---|
| | < 100[a] | 100–300 | 300–1000 | 1000–3000 | 3000–5000 | > 5000 |
| 25–1000 | 187 | 292 | 193 | 64 | 21 | 21 |
| 1001–5000 | 59 | 44 | 32 | 3 | 0 | 1 |
| 5001–50000 | 19 | 26 | 11 | 0 | 0 | 0 |
| > 50,000 | 2 | 1 | 0 | 0 | 0 | 0 |
| Totals | 267 | 363 | 236 | 67 | 21 | 22 |

Note: Population-weighted mean = 648 pCi/L, assuming that the concentration below the minimum reporting level = 0.5 pCi/L.
[a]Minimum reporting level.

## HEALTH EFFECTS AND RISK FROM RADIONUCLIDES IN DRINKING WATER

### Radium

Radium, biochemically similar to barium and calcium when ingested, concentrates in the bone. The health effects of ingested radium are well documented and are largely based upon studies of radium watch dial painters who worked in the early part of this century. These women ingested large amounts of radium when the brushes they were using to apply radium-containing fluorescent paint to watch faces were "pointed" with their tongues, teeth, and lips. In the late 1920s and 1930s, bone sarcomas and head carcinomas were diagnosed among this group, but no statistically significant number of leukemias have been found. The total number of combined leukemias and bone cancers predicted from the existing dosimetric model is very nearly the same as the numbers of predicted bone cancers based on the study of the watch dial painters. Ingesting 5 pCi/L of radium in water, at 2 L per day for 70 years, gives an expected risk rate of $4 \times 10^{-5}$. (This estimate and those in the rest of this chapter assume a linear no-threshold dose-response curve.) Additionally, since radium is distributed throughout the volume of the bone, one might expect that the red bone marrow would be exposed and some leukemias would result from this ingestion. However, for reasons not understood, none of the radium watch dial painters' cohort has shown a statistically significant level of leukemia.

### Uranium

Ingested uranium goes primarily to the bone and to the kidney. Natural uranium's major isotopes (uranium-235 and uranium-238) have exceptionally low radioactivity per gram of material (a consequence of their long half-lives). This ratio, known as the specific activity, is roughly a million times lower than that of the radium isotopes. Because of natural uranium's low specific activity, it is not experimentally possible to dose animals with natural uranium (which is 0.006% uranium-234, 0.72% uranium-235, and 99.27% uranium-238) such that the total radioactive dose delivered to the bone will induce cancers while at the same time avoiding toxic life-shortening chemical and physiological reactions (e.g., nephritis) to the kidney. An experiment by Finkel used the comparatively short-lived (high specific activity) manmade uranium isotopes $^{232}U$ and $^{233}U$ and successfully demonstrated that the alpha particles emitted from the uranium nucleus are not qualitatively different in terms of their carcinogenic potential from alphas emitted by the known radionuclide carcinogens radon and radium.[11] This limited evidence, with the presumption that all energetic alpha particles are carcinogenic when they are absorbed in the bone, suggests the use of the health effects data for radium as a surrogate for natural uranium since both species bind to the bone and both emit alpha particles during

radioactive decay. Uranium is also known to have deleterious or chemotoxic effects to the kidney.[12] In estimating the risk due to uranium in drinking water, the crucial questions are the following:

1. How much ingested uranium goes to the bone, and how much goes to the kidney?
2. What are their relative consequences?

The International Commission on Radiation Protection (ICRP) estimates that an average of 5% of ingested uranium is absorbed by the bloodstream from the gastrointestinal tract. It also suggests that the value could possibly be as high as 20%, as had been shown in some earlier work.[12,13] At the National Workshop on Radionuclides in Drinking Water in 1983, a consensus value for gastrointestinal absorption of 1.4% was suggested.[14,15] Recent experiments yield a better measurement of this uptake value and its determinants.[16] Preliminary results from experiments that used fasting human subjects suggest a value in the 0.5 to 1.0% range. In view of these results, both the 1.4% and the 5% estimates arguably represent reasonable, and at the same time prudent, average levels to use in risk calculations.

### Radon

Inhaled radon has been shown in several epidemiologic studies to lead to lung cancer. Studies have been made of uranium miners in Colorado, Canada, and Czechoslovakia, hard rock miners in Sweden, and fluorspar miners from Newfoundland. These studies involve a number of different ethnic types, different smoking habits, and different mining conditions. The range of uncertainty in the resulting risk factors is no more than an order of magnitude when extrapolated to environmental concentrations. The dose-response relationship from these epidemiological studies is also well supported by data from animal studies on rats and mice.[4]

Recent experimental work related to the ingestion of radon has been analyzed.[17,18] Although the uncertainty is large, this work suggests the number of fatal cancers from radon ingested from drinking water may be equal to a significant fraction of the fatal lung cancers due to the radon inhaled from that portion of radon in indoor air that originates from the off-gassing of radon from drinking water. (See also Chapter 2.)

## ESTIMATED RISK DUE TO NATURALLY OCCURRING RADIONUCLIDES IN DRINKING WATER

Table 6 shows the estimated risk levels (using the linear no-threshold dose-response curve hypothesis) due to different concentrations in drinking water of the naturally occurring radionuclides, calculated using weighting factors developed by the ICRP. The risk levels shown range from one in a thousand to one in a million per lifetime. Also shown are the corresponding annual effective

**Table 6. Lifetime Risks for Drinking Water Radionuclides**

| Risk | Dose (mrem/yr) | Concentration | | | |
|---|---|---|---|---|---|
| | | Ra-226 | Ra-228 | Nat U | Rn-222 |
| $10^{-3}$ | 300 | 100 | 100 | 600 | 2000 |
| $10^{-4}$ | 30 | 10 | 10 | 60 | 200 |
| $10^{-5}$ | 3 | 1 | 1 | 6 | 20 |
| $10^{-6}$ | 0.3 | 0.1 | 0.1 | 0.6 | 2 |

Note: Data are shown to one significant figure.

**Table 7. Mean U.S. Groundwater Concentrations of Radionuclides and Associated Lifetime Risks**

| Radionuclide | Mean Concentration (pCi/L) | Lifetime Risk |
|---|---|---|
| Radium-226 and -228 | 1.1 | $1 \times 10^{-5}$ |
| Radon | 600 | $4 \times 10^{-4}$ |
| Natural uranium | 1.2 | $2 \times 10^{-6}$ |

dose equivalents in mrems/year. The mean concentration in U.S. public drinking water supplies (served by groundwater and weighted for the populations they serve) is shown with its associated lifetime risks in Table 7.

Table 8 presents the number of deaths that can be attributed to each radionuclide during a 70-year interval. Clearly, radon dominates the mortality data and is responsible for about 80% of the radionuclide-induced deaths.

## POSSIBLE MAXIMUM CONTAMINANT LEVEL STANDARDS FOR RADIONUCLIDES IN DRINKING WATER

The Safe Drinking Water Act directs the U.S. EPA to establish primary drinking water regulations for public water systems, to specify contaminants judged to pose an adverse effect on human health, and to specify the maximum contaminant level for each of these designated contaminants or alternatively, to specify an appropriate treatment technique to mitigate each contaminant. Interim primary regulations were to be established less than 180 days after publication of the regulations in the *Federal Register* on July 9, 1976. These regulations were developed in two steps: first, recommended maximum contaminant levels were to be proposed and promulgated, followed by the

**Table 8. Number of Deaths in Seventy Years (Annually) from Radionuclides in Drinking Water**

| Isotope | Range |
|---|---|
| Ra-226 | 200—4000 (3–60) |
| Ra-228 | 200—4000 (3–60) |
| U | 100—1000 (2–20) |
| Rn-222 | 6000—60,000 (80–800) |
| Sr-90 | 10—70 (0.1–1) |

Note: All values have been given to one significant place.

**Table 9. Maximum Contaminant Levels as Set by the 1976 National Interim Regulations**

| | |
|---|---|
| Gross alpha[a] | 15 pCi/L |
| Radium-226 and -228 | 5 pCi/L |
| Gross beta | 50 pCi/L |
| Manmade radionuclides | 4 mrem |

[a]Exclusive of contribution from uranium and radon, for which insufficient data were available to set regulations.

proposal and issuance of the maximum contaminant levels. The radionuclides found in water are proven (or, in the case of natural uranium, very strongly inferred) human carcinogens. Carcinogens are considered to have no dose below which adverse effects occur (e.g., no threshold).

An analysis of the health risks and feasibility of remediation led the U.S. EPA to set the interim regulations as shown in Table 9. To have set these guidelines in 1976 required a balancing process that considered several independent but related factors.

Figure 1 shows the number of fatalities averted for each of three major naturally occurring radionuclides as a function of setting maximum concentration limits for the country's public drinking water supplies. The figure has been drawn based upon the geometric mean risk values for each nuclide as listed in

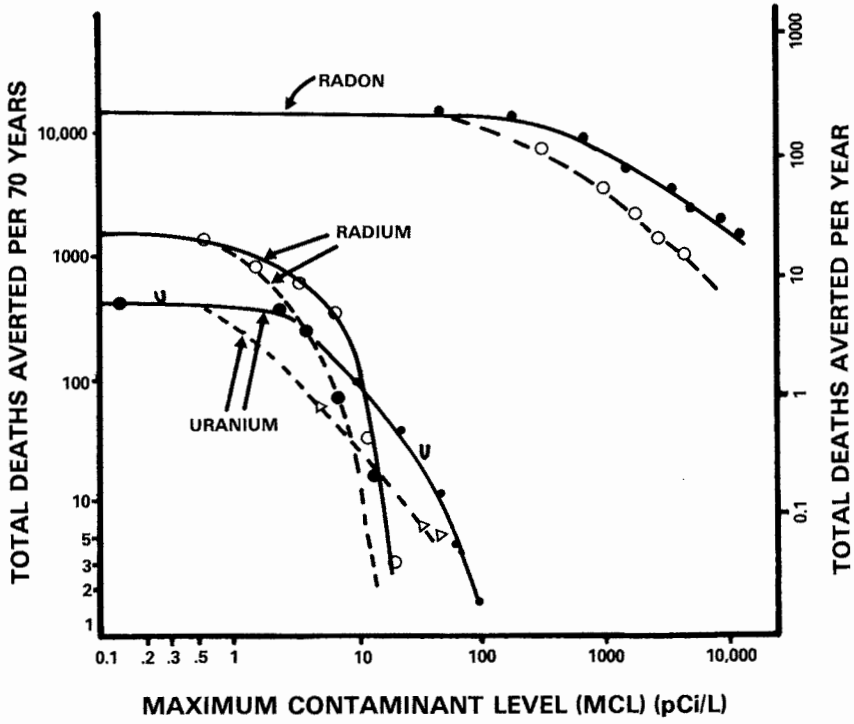


**Figure 1.** Total deaths averted in a lifetime of seventy years and annually as a function of a range of possible maximum contaminant levels.

Table 8. Other assumptions made are that the average radium and uranium concentrations in surface water supplies are the same as in groundwater supplies and that radon surface water concentrations are zero. For clarity of presentation, the remainder of the text also presents the number of deaths or deaths prevented using the geometric mean rounded to one significant figure. By selecting a proposed regulatory maximum contaminant level along the abscissa, the number of cancer fatalities averted annually or during a 70-year interval may be read on the ordinate. For example, the number of deaths from exposure to radium from the approximately 500 water supplies not in compliance (above the 5 pCi/L radium maximum contaminant level) equals a total of about 1500 fatalities over 70 years (or about 7 annually). By bringing these water supplies into compliance, about 200 fatalities would be prevented, which is about 15% of the total radium-induced fatalities.

No interim maximum contaminant level standard for uranium in drinking water exists at this time. Based upon both chemotoxicity and radiological risk, many experts have suggested that the maximum contaminant level be set in the range of 20 to 100 pCi/L. Based upon the NIRS survey, less than 4% of the nation's 60,000 public water supplies exceed the lower value and less than 0.5% exceed the higher value.[19] From Figure 1 we observe that setting a 20 pCi/L level (representing a lifetime risk of $4 \times 10^{-5}$) would avert about 3% of the uranium-induced fatal cancers due to the drinking water source.

Radon is the radionuclide that represents the largest risk of all the radionuclides in drinking water. Figure 1 shows that about 17,000 fatal cancers occur over a 70-year interval from exposure to radon originally present in drinking water. Setting the maximum contaminant level at the population-weighted national average groundwater concentration of about 600 pCi/L would prevent about 30% of the radon fatalities. About one-half of the public drinking water supplies that use groundwater would exceed this level and would require remediation. This, in turn, requires a very substantial effort (which can, in turn, be measured in dollars) and achieves a maximum radon risk ($4 \times 10^{-4}$) that exceeds the ballpark range of what some consider an acceptable risk of $10^{-4}$ or less. An alternative maximum contaminant level of 2000 pCi/L represents a lifetime cancer risk of $1 \times 10^{3}$ and would require about 8% of public water supplies that have radon concentrations greater than this concentration to remediate their water. This would avert about 30 annual cancer deaths. Clearly, this maximum contaminant level is less protective of health but, as will be shown below, is considerably less expensive to implement.

Based upon these examples, it is relatively straightforward to select reasonable ranges of radionuclide maximum contaminant levels for each of the radionuclides. But to fine-tune this range and select a single, specific maximum contaminant level that is consistent with health considerations and the legal strictures of the Safe Drinking Water Act (which allow "taking cost into account") is considerably more difficult.

A more comprehensive and less subjective approach to setting maximum contaminant levels for the radionuclides in public drinking water supplies

**Table 10. Comparison of Total Radium, Uranium, and Radon Activities Based upon Nine Variables**

| | Variable | Radium-226 + -228 | Radon-222 | Uranium |
|---|---|---|---|---|
| 1. | 4.5 × average concentration | 5 | 3000 | 6 |
| 2. | $4 \times 10^{-5}$ lifetime risk | 5 | 60 | 20 |
| 3. | 15% of total deaths averted | 5 | 1000 | 5 |
| 4. | 3 deaths averted annually | 5 | 10000 | 2 |
| 5. | 5 mrem/year | 5 | 200 | 30 |
| 6. | 1% not in compliance | 5 | 10000 | 30 |
| 7. | 8 × minimum reporting level | 5 | 800 | 0.6 |
| 8. | \$3 million/year = cost to avert 1 fatality for radium MCL = 5 pCi/L | 5 | 200 | 70000 |
| 9. | \$9 million/year = total remediation cost for radium MCL = 5 pCi/L | 5 | 4000 | 100 |
| | Geometric mean of variables 1–9 | 5 | 1000 | 30 |

Note: All concentrations in 5pCi/L.

might utilize the basic inputs (independent variables) that helped in the selection of the 1967 radium maximum contaminant level. The 5 pCi/L maximum contaminant level for this radionuclide has stood the test of time. Based upon Figure 1 and the 5 pCi/L maximum contaminant level already established for total radium, the U.S. EPA may consider possible uranium and radon maximum contaminant levels. The radium concentration of 5 pCi/L limits the risk to less than $4 \times 10^{-5}$ (that is, all public drinking water supplies above this risk are deemed not to be in compliance and must be remediated). The maximum contaminant level for radium is also about 4.5 times the U.S. population-weighted average radium concentration. The 5 pCi/L radium maximum contaminant level would avert (when all public drinking water supplies not in compliance remediate their systems by reducing the radium concentration to the maximum contaminant level) about 15% of the total cancer fatalities caused by radium in drinking water. Finally, about 1% of the public drinking water supplies have radium concentrations higher than the maximum contaminant level and require remediation. Based upon the radium example, Table 10 (variables 1 through 7) provides maximum contaminant levels for uranium and radon for these and additional independent variables. For radon they vary by a factor of nearly 200, and for uranium by a factor of 120,000.

Figure 2 provides comparative cost data that may also be considered when examining alternative maximum contaminant level values.[20] By referring to this figure, for any selected maximum contaminant level, the average cost (costs for smaller and larger water supplies have been aggregated) to prevent a cancer death is shown. At lower maximum contaminant levels, although more cancer deaths are prevented, the costs become disproportionally larger so that the cost per cancer prevented increases as the maximum contaminant level is reduced.

The costs of remediation per cancer case prevented is shown on line 8 of

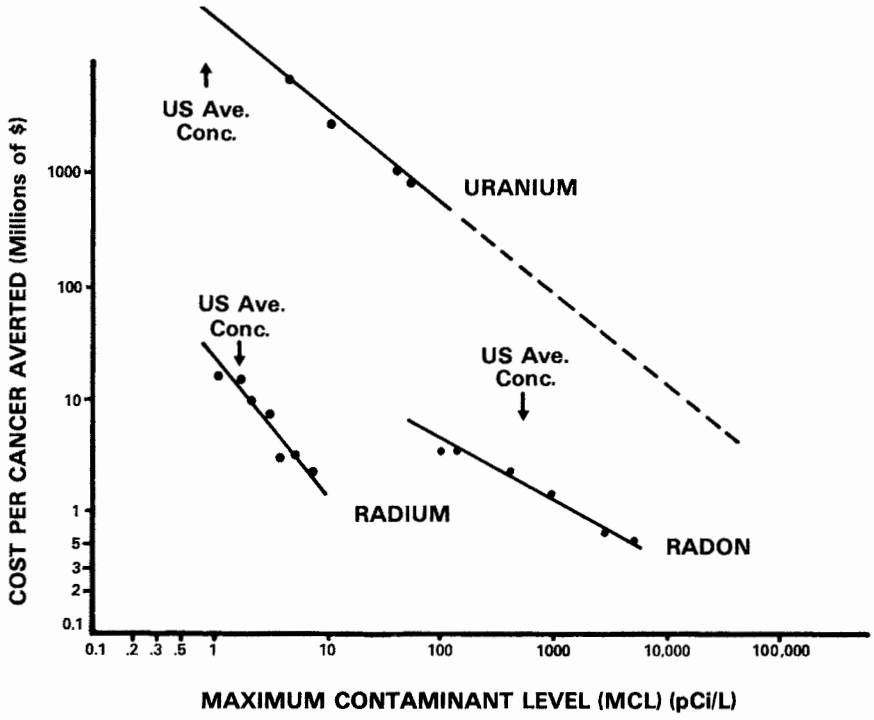


**Figure 2.** Cost per cancer averted for a range of maximum contaminant levels of uranium, radium, and radon.

Table 10, using the radium maximum contaminant level of 5 pCi/L as the point of reference. Reducing radium in all water supplies to this value would cost about $3 million per cancer death prevented (see Figure 2). From Figure 2 it is also seen that the same cost to prevent a cancer death sets the uranium and radon maximum contaminant levels at about 70,000 pCi/L and 200 pCi/L, respectively. For uranium this maximum contaminant level is meaningless, since no public water supply has been found to have concentrations higher than 600 pCi/L. Thus, setting a 600 pCi/L maximum contaminant level for uranium is equivalent to not regulating uranium. In the absence of any regulations to reduce uranium in public drinking water supplies, about 6 deaths per year would result from cancer induced by uranium exposure, a lifetime risk equal to $2 \times 10^{-6}$.

Another approach considers the total or national costs to reduce the radionuclide water concentrations to the maximum contaminant level values. For example, to bring into compliance those public water supplies whose water presently exceeds the 5 pCi/L for radium would cost about $9 million per year (annualized capital costs discounted at 3% over 20 years plus monitoring and operating costs in 1986 dollars), since Figure 1 indicates 5 pCi/L would prevent about 3 cases per year and Figure 2 indicates the cost per death prevented

would be about $3 million. This includes disposal costs at sanitary waste disposal facilities for the radioactive species that have been extracted from the water. Disposal at low-level radioactive waste disposal sites would be more expensive by approximately a factor of five. Table 10, line 9 indicates that $9 million would also allow all public water supplies with radon concentrations above 4000 pCi/L to remediate for this radionuclide. Similarly, for this total annual cost, all uranium concentrations could be reduced to about 100 pCi/L.

Nine different criteria for selecting the uranium and radon maximum contaminant levels are shown in Table 10, all based upon the corresponding radium value for each criterion. The table shows that there are wide variations in the values of these variables. However, line 10 of Table 10 presents the geometric mean for each radionuclide of the concentrations corresponding to variables 1 through 9. These values for radon and uranium both fall, perhaps fortuitously, within the seemingly reasonable ranges of radionuclide maximum contaminant levels discussed earlier in this section.

An alternative regulatory approach would not set the individual maximum contaminant levels based solely on their individual risk (as modified by feasibility and related considerations). Rather, for example, it might emphasize the overall health aspects of the problem. This approach would dictate that available resources be used so that maximum health benefits accrue. It is evident that by regulating uranium less stringently the resources saved could then be used to reduce further the radon maximum contaminant level to a concentration lower than would otherwise seem practicable. Thus (using Figure 3, which is derived from Figures 1 and 2) a uranium maximum contaminant level equal to 100 pCi/L would cost about $60 million per year, essentially $240 million less than the cost of regulating at a 23 pCi/L uranium value. Such a relatively high uranium standard barely achieves any protection of health (it would prevent about 0.05 of the 6 annual uranium deaths, although the 23 pCi/L standard itself prevents no more than 0.2 death). But the uranium concentrations that are seen in public drinking water supplies represent risks that already, prior to any regulatory effort, can be arguably considered acceptably low.

A very substantial improvement from an overall risk perspective can be achieved by setting the uranium and radon maximum contaminant levels in consort so that they maximize cancer deaths prevented per dollar expended. Let us examine two examples. The first starts by assuming a radon maximum contaminant level of 2000 pCi/L and the second a standard of 500 pCi/L. By utilizing the $240 million per year that would be realized by not setting the uranium standard at 23 pCi/L but rather at the 50 pCi/L concentration, more stringent control of radon can be effected.

Figure 3 shows the annual cost of setting various uranium and radon maximum contaminant levels. As has been seen, the difference in cost between implementing a uranium standard of 23 pCi/L and one equal to 50 pCi/L is approximately $240 million. If a risk manager had concluded that 2000 pCi/L and 23 pCi/L were appropriate maximum contaminant levels for radon and

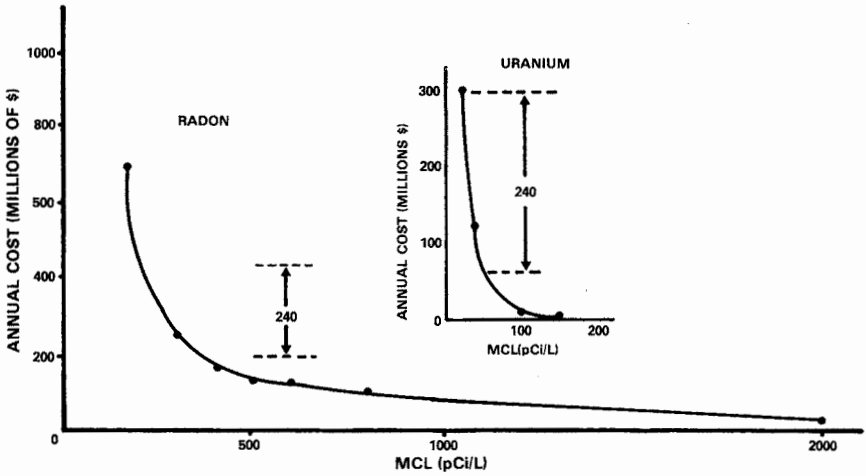


**Figure 3.** Costs for a range of maximum contaminant levels possibilities.

uranium considered separately, by raising the uranium from 23 pCi/L to 50 pCi/L (at a health cost of less than 0.2 additional deaths per year and a saving in resources of $240 million), he or she could apply these resources to reduce the radon maximum contaminant level from the postulated 2000 pCi/L to 280 pCi/L. This would prevent an additional 90 radon deaths annually, or a total of 120 total annual deaths. Similarly, were the radon maximum contaminant level initially selected to be 500 pCi/L, $240 million enables, at no extra cost, but by virtue of using the resources "saved" by raising the uranium maximum contaminant level to 50 pCi/L, the radon maximum contaminant level to be set at about 230 pCi/L. Compared to the 500 pCi/L, this prevents an additional 40 deaths to give a total of 130 annual deaths.

In view of these examples, and recognizing that by virtue of raising the uranium maximum contaminant level to 50 pCi/L, a very small number (about 0.1) of additional deaths per year would not be prevented, it is clear that the net result is a substantial improvement in overall risk reduction. Finally, it should be observed that placing the uranium maximum contaminant level at 50 pCi/L is equivalent to allowing public drinking water supplies to pose a maximum lifetime uranium risk equal to $1 \times 10^{-4}$. The radon maximum contaminant levels of 210 pCi/L and 260 pCi/L are equivalent to lifetime risks of $1.3 \times 10^{-4}$ and $1.6 \times 10^{-4}$, marginally higher than the previously mentioned $10^{-4}$ "acceptable" level. The overall reduction of national average radon lifetime risk from public ground and surface water supplies is not a trivial reduction as it prevents 50% of the more than 200 annual radon deaths. Since, relatively, so few supplies exceed the suggested uranium MCLs, the average uranium risk prior to and after setting a maximum contaminant level of 23 pCi/L or 50 pCi/L will remain essentially unchanged at $2 \times 10^{-6}$.

## CONCLUSION

We have examined the occurrence, exposure, and risks posed by naturally occurring radionuclides in public drinking water supplies. In the context of these data and considering the feasibility and cost of compliance, we have considered several regulatory approaches that could be implemented to protect public health. We have examined hypothetical, but realistic, maximum contaminant levels for radon and uranium and their effect on reducing the health risks of drinking water that these two radionuclides pose. We suggest that the flexibility of the Safe Drinking Water Act allows setting of the maximum contaminant levels for the regulation of radon's and uranium's alpha particle effects at levels that are not totally independent of each other, but that are interrelated such that maximum health benefits are achieved. To achieve this, as we have sought to illustrate, requires a policy that emphasizes a relatively stringent radon standard. It can be achieved by relaxing the uranium standard at little sacrifice to health, but with a large saving of resources. To justify this, these resources must then be used to reduce the considerably greater risk posed by radon. A net reduction in risk is achieved at no increased resource expenditure. We note, too, that neither the uranium nor the radon maximum contaminant level is set at a level that significantly exceeds the level of risk considered by the U.S. Environmental Protection Agency as being protective of public health.

## NOTICE

The thoughts and ideas expressed in this chapter are those of the authors and are not necessarily those of the U.S. Environmental Protection Agency.

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# CHAPTER 2

# Analysis of the Health Risk from Ingested Radon

**Douglas J. Crawford-Brown**

## INTRODUCTION

The human population typically is exposed to radiation from a number of sources, both internal and external. These radiations arise primarily from the decay of potassium and ruthenium isotopes, as well as from the members of the uranium, thorium, and actinium decay chains. In addition, significant doses are delivered as a result of cosmic radiation arriving at the surface of the earth. Attention in the risk analysis community recently has been shifted from these sources, however, because of findings that geological sources of $^{222}Rn$ (hereafter, radon) may produce risks larger than those produced by the other sources. A comparison of annual dose equivalent received from radon and other environmental sources reveals that radon in the air of homes yields roughly 20–30 mSv per year (with a quality factor of 20) in the United States,[1] whereas the others yield less than 1 mSv per year.[2] Even if the whole body effective dose equivalent, rather than the dose equivalent alone, is used as the index of damage, radon yields 2 to 3 times the value noted for other radionuclides in the environment.

Previous risk assessments for radon have examined the exposures that occur from radon in the air.[1] It usually is assumed that airborne radon is a much greater hazard than ingested radon in daily life. As a result, even risk assessments specific to radon in water supplies have focused on the emanation of the radon from the water and into the air of the home.[3,4] The assumption is that the risk imposed by the use of water containing radon may be approximated by the inhalation route, with little need to include the risk from direct ingestion. The purpose of this chapter is to explore this assumption by calculating the lifetime risk imposed by direct ingestion of radon in water.

As demonstrated in Figure 1, the concentration of radon in drinking water obtained from public supplies varies widely. In the present chapter, it will be assumed that the population-weighted average concentration of radon in U.S. public drinking water supplies is 4 Bq of radon per liter of water.[4] It should be mentioned, however, that the concentration of radon in groundwater supplies can greatly exceed this average value. Since a linear dose-response function for

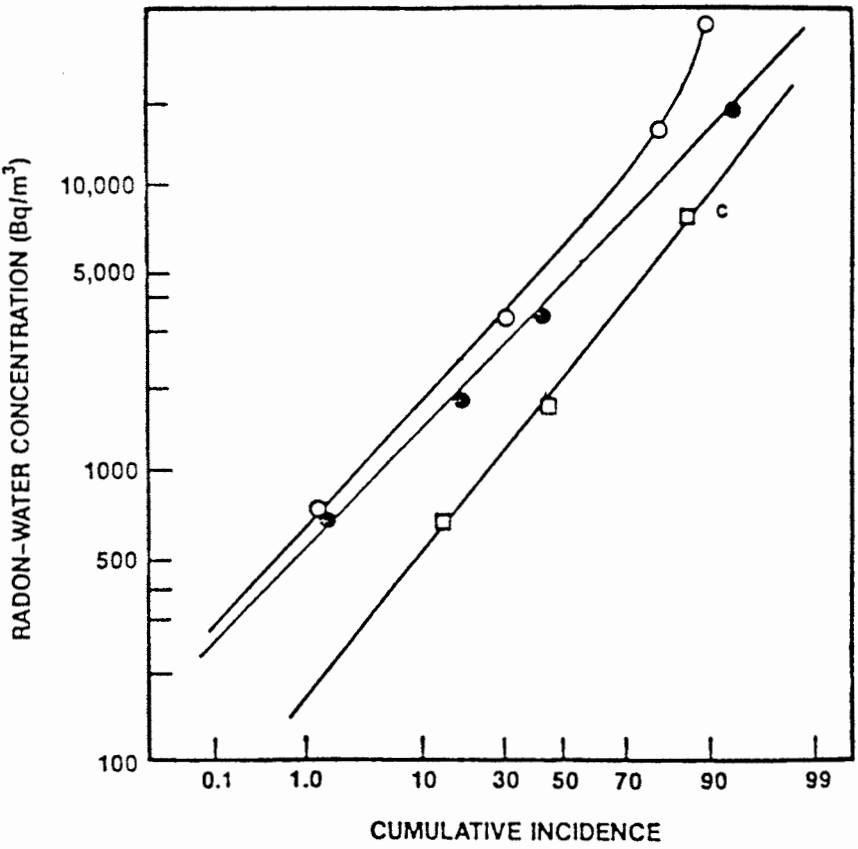


**Figure 1.** Distributions of radon concentration for the United States: *a*, in public groundwater supplies; *b*, in public groundwater supplies weighted by population served by each supply; *c*, in public and private groundwater supplies weighted by population served by each supply. The horizontal scale is the percent that exceed that concentration.

cancer induction will be adopted, the lifetime risk for any other concentration of radon may be obtained by scaling linearly.

## EARLY STUDIES

Interest in the biokinetics of radon in the human body began primarily in the 1960s. These studies, performed using human volunteers, involved ingestion of water containing known quantities of radon. The movement of the radon through the body then was followed, allowing limited estimation of whole body doses and doses to selected tissues.

Andersson and Nilsson[5] used whole body counting to study the retention of radon in the whole body. Their results indicated that the radon was retained in

the body with a half-time of between 30 and 70 minutes, although a smaller long-term half-time of several hundred minutes also was noted. Their results were substantiated by later studies.[6-9] From their study, it may be estimated that the dose equivalent to the whole body (with a quality factor of 20) for adults was approximately $4 \times 10^{-9}$ Sv per Bq of ingested radon. This value assumes a quality factor of 20 for the radiobiologically significant alphas,[10] which will be used throughout this report. Doses to the stomach probably were higher, and other tissue doses probably were lower, than their reported value, on account of their inability to separate the two components from the measurement data.

Von Dobeln and Lindell[7] conducted a similar study and found a whole body dose equivalent of approximately $1 \times 10^{-9}$ Sv per Bq of ingested radon. Again, since whole body imaging was used, doses to separate tissues could not be calculated. Hursh et al.,[8] however, conducted a more extensive study that examined separate compartments of the body, including venous blood and exhaled air. From this study, it may be concluded that doses generally will be lowest for systemic body tissues, with the perfused liver and the gastrointestinal (GI) tract (particularly the stomach) receiving substantially higher doses. Their estimate of the whole body dose is similar to those cited above, with the liver being higher by a factor of 10 due to its perfusion with blood arriving directly from the GI tract. In addition, the Hursh et al.[8] study demonstrated that the radon equilibrates quickly with the liver and portal blood, since the maximum concentration in the liver is established in 5 to 15 minutes following ingestion.

This same study also demonstrated that ingested radon leaves the body primarily through exhalation via the lung: greater than 95% of the radon leaves by this route. As demonstrated by Suomela and Kahlos,[9] this pattern holds true for humans and rats, indicating that the primacy of this route and the half-time of removal are related only to the ratio of lung surface area to tissue mass, which is similar in the two species. This finding has been used by Crawford-Brown[11] to suggest that the removal half-time and route of removal also are independent of age in humans ingesting radon.

It generally is assumed that doses to fatty tissues will be higher than those to other systemic tissues. This assumption arises from the finding of Nussbaum and Hursh[12] that the solubility coefficient of radon in fat is increased significantly above that for water. The suggestion may be made, therefore, that any long-term retention of radon in the body may be dominated by the content in fatty tissue. This assumption has not been demonstrated to date.

## BIOKINETICS MODELS

The most detailed organ-specific measurements of radon have been performed by Correia et al.[13] at Massachusetts General Hospital as part of the U.S. Environmental Protection Agency (EPA) effort to regulate radon. These