

UNC-WRRI-91-260

USE OF ACTIVATED CARBON
TO REMOVE RADON FROM DRINKING WATER

by
James E. Watson, Jr.
and
Douglas J. Crawford-Brown

Department of Environmental Sciences and Engineering
University of North Carolina
Chapel Hill, North Carolina 27599-7400

The research on which the report is based was financed in part by the United States Department of the Interior, Geological Survey, through the N.C. Water Resources Research Institute.

Contents of the publication do not necessarily reflect the views and policies of the United States Department of the Interior, nor does mention of trade names or commercial products constitute their endorsement by the United States Government.

Agreement No. 14-08-0001-G1580
WRRI Project No. 70099
USGS Project No. 06 (FY89)

ABSTRACT

The U.S. Environmental Protection Agency has announced its intent to issue a standard for radon (^{222}Rn) in public drinking water supplies and it is expected that many drinking water sources in North Carolina will require radon removal. This project studied the feasibility of using granular activated carbon (GAC) as a point-of-entry treatment method to remove radon from individual drinking water supplies. Aeration is the recommended method for treatment of public water supplies.

When radon is adsorbed on GAC, it decays with a half-life of 3.8 days to produce a series of radioactive decay products. The first 4 decay products of radon have short half-lives - all less than 30 minutes. The fifth decay product in this series is ^{210}Pb which has a half-life of 22 years. The build-up of ^{210}Pb could require the GAC to be disposed of as radioactive waste.

At the present time there are no federal regulations specifically applicable to the disposal of GAC containing natural radionuclides. EPA is currently considering the development of regulations controlling the disposal of waste containing naturally occurring radionuclides in excess of a concentration of 2,000 pCi/g. EPA has also recently published "suggested guidelines" for the disposal of naturally occurring radionuclides generated by drinking water treatment plants that recommend restrictions on disposal of waste with substantially lower concentrations of ^{210}Pb .

This project included an experimental determination of the removal fraction for radon from drinking water by GAC. The experimental design consisted of flowing radon laden water through a GAC column, measuring the inlet and outlet concentrations of radon in the water, and measuring the activity of ^{210}Pb in slices of GAC from the column. The results of this work indicated a removal fraction of radon from drinking water of approximately 0.95. The results also indicated that nearly 100% of the ^{210}Pb produced from the decay of the radon was retained on the GAC.

Calculations were performed of the build-up of ^{210}Pb on GAC used to remove radon from water. The build-up of ^{210}Pb is a function of the radon concentration, water use rate, radon removal efficiency, decay product retention, and length of exposure of the GAC to radon. Assuming a 15 kg mass of GAC, a radon concentration of 10,000 pCi/l, use rate of 1,000 l/day, 95% removal efficiency, and 100% decay product retention, it was calculated to take over 25 years for the GAC to reach a concentration of 2,000 pCi/g. Thus, for these conditions, and a limit of 2,000 pCi/g, GAC could be used for radon removal without becoming low-level radioactive waste if it were replaced within approximately 25 years. For higher radon concentrations and/or larger use rates, the replacement of GAC would be required at more frequent intervals. However, the majority of individual water sources in North Carolina, that are likely to exceed the standard for public water supplies, have radon concentrations less than 10,000 pCi/l.

It appears that GAC systems could be used to remove radon from most individual North Carolina water supplies without the GAC being classified as low-level radioactive waste (based on a criterion of 2,000 pCi/g) if the GAC occasionally is replaced. More restrictive requirements on the disposal of solid waste containing natural radionuclides would limit the feasibility of using GAC to remove radon from drinking water.

TABLE OF CONTENTS

	PAGE
Abstract	ii
List of Figures	iv
List of Tables	v
Summary and Conclusions	vi
Recommendations	viii
Introduction	1
Disposal Requirements	4
Computations	6
Experimental Methods	12
Experimental Results	17
References	20

LIST OF FIGURES

	Page
1. Build-up of ^{210}Pb on GAC for ^{222}Rn Water Concentration of 10^4 pCi/l and Water Use Rate of 10^3 l/d	8
2. Time at Which ^{210}Pb Concentration on GAC Will Equal 2,000 pCi/g	10
3. Time at Which ^{210}Pb Concentration on GAC Will Equal 50 pCi/g	11
4. System for Experimental Determination of Radon Removal from Water	13

LIST OF TABLES

	Page
1. Measurement Results for NaI Analysis	18
2. Water Flow Rate and Radon Concentration Measurements	18
3. Raw Data Obtained for the Inlet and Outlet Water Measurements	19
4. Half-time Determination for Inlet Water Sample Obtained on 9/9/90	19

SUMMARY AND CONCLUSIONS

The U.S. Environmental Protection Agency (EPA) has announced its intent to issue a standard for radon (^{222}Rn) in public drinking water supplies. Since half the population of North Carolina relies on groundwater for drinking water and since studies indicate that the radon content of N.C. groundwater supplies may be significantly higher than the U.S. average, it is expected that many drinking water supplies, including many individual supplies, in the state will require radon removal. Individual drinking water sources, which will not be regulated by the EPA standard but which should be treated, have some of the highest radon concentrations.

Two principal methods have been proposed for removing radon from drinking water: aeration and adsorption on granular activated carbon (GAC). Aeration is the recommended method for treatment of public water supplies. Because, for point-of-entry applications, aeration is expensive and requires more maintenance, it is expected that GAC removal may be better suited for individual wells. However, questions about radiation exposure resulting from build-up of radioactivity on the GAC and disposal of GAC have not been resolved. This project addressed the feasibility of using GAC to remove radon from drinking water.

When ^{222}Rn is adsorbed on GAC, it decays with a half-life of 3.8 days to produce a series of radioactive decay products. The first four decay products of radon have short half-lives all less than 30 minutes. (Radon and its short-lived decay products produce a radiation field near the GAC during its use. This exposure can be minimized by either locating the GAC where it will not be a problem or by shielding the system.) The fifth decay product in the series from radon is ^{210}Pb which has a half-life of 22 years. The build-up of ^{210}Pb could require that the GAC be disposed of as radioactive waste.

At the present time there are no federal regulations specifically applicable to the disposal of GAC containing natural radionuclides. A survey was conducted of states with operational low-level radioactive waste disposal sites or with plans to develop disposal sites to determine whether these states have regulations applicable to the disposal of GAC containing natural radionuclides. This survey revealed that most states do not have pertinent regulations. Many states responded that they will follow guidance from the U.S. Environmental Protection Agency. EPA is currently considering the development of regulations controlling the disposal of wastes containing naturally occurring radionuclides in excess of a concentration of 2,000 pCi/g. EPA also recently published "suggested guidelines" for the disposal of naturally occurring radionuclides generated by drinking water treatment plants. These guidelines recommend restrictions on the disposal of solid wastes containing as little as 3 pCi/g of ^{210}Pb and recommend consideration of disposal in RCRA permitted hazardous waste units for wastes with greater than 50 pCi/g of ^{210}Pb . The State of Washington already has regulations requiring that wastes containing greater than 2,000 pCi/g of natural radionuclides be disposed of as low-level radioactive waste.

This project included an experimental determination of the removal fraction for radon from drinking water by GAC. A source of radon-laden water was produced in the laboratory using a radon generator. The experimental design consisted of flowing the water through a GAC column, measuring the inlet and outlet concentrations of radon in the water, and measuring the activity of ^{210}Pb in slices of GAC from the column. The concentration of radon in water at both the inlet and the outlet was determined by liquid scintillation counting of the alpha emissions from the radon decay chain at equilibrium. Measurement of the ^{210}Pb activity on the GAC was accomplished by counting the photon emissions using a NaI spectrometry system. The results

of this work indicated a removal fraction for radon from drinking water of approximately 0.95. The results also indicated that nearly 100% of the ^{210}Pb produced from the decay of the radon was retained on the GAC.

Calculations, using the Bateman Equations, were performed of the build-up of ^{210}Pb on GAC used to remove radon from water. The build-up of ^{210}Pb is a function of the radon concentration, water use rate, radon removal efficiency, decay product retention, and length of exposure of the GAC to radon. The concentration of ^{210}Pb on GAC of course depends on the mass of GAC on which the ^{210}Pb is adsorbed. Assuming a 15 kg mass of GAC, a radon concentration of 10,000 pCi/l, a use rate of 1,000 l/day, 95% removal efficiency, and 100% decay product retention, it was calculated to take over 25 years for the GAC to reach a concentration of 2,000 pCi/g. For these same conditions the concentration will exceed 50 pCi/g in less than 1 year.

It appears likely that GAC containing greater than 2,000 pCi/g of natural radionuclides will require disposal as low-level radioactive waste. Restrictions may be placed on disposal of GAC with lower radionuclide concentrations. It was determined that for water with a radon concentration of 10,000 pCi/l and a use rate of 1,000 l/day, the activity of ^{210}Pb on the GAC will not reach 2,000 pCi/g for approximately 25 years. Thus for these conditions, and a limit of 2,000 pCi/g, GAC could be used for radon removal without becoming low-level radioactive waste if it were replaced within approximately 25 years. For higher radon concentrations and/or larger use rates, the replacement of GAC would be required at more frequent intervals. However, the majority of individual water sources in North Carolina that are likely to exceed the standard for public supplies, have radon concentrations less than 10,000 pCi/l. It appears that GAC systems could be used to remove radon from these water supplies without the GAC being classified as low-level radioactive waste if the GAC occasionally is replaced. More restrictive requirements on the disposal of solid wastes containing natural radionuclides would limit the feasibility of using GAC to remove radon from drinking water.

RECOMMENDATIONS

GAC may be better suited for removing radon from individual drinking water supplies than aeration because aeration is more expensive and aeration systems require more maintenance. The most significant factor in determining the feasibility of using GAC to remove radon from drinking water is likely to be the matter of whether the GAC becomes low-level radioactive waste. The EPA has announced that it is considering the development of regulations controlling the disposal of waste containing naturally occurring radionuclides in excess of a concentration of 2,000 pCi/g, but has also published "suggested guidelines" for the disposal of naturally occurring radionuclides generated by drinking water treatment plants that contain recommended restrictions for waste with substantially lower concentration of ^{210}Pb . If waste containing less than 2,000 pCi/g of ^{210}Pb can be disposed of as ordinary waste, it appears that GAC systems could be used to remove radon from the majority of individual water sources in North Carolina that are likely to exceed the standard for public supplies. However, more restrictive requirements on the disposal of wastes containing ^{210}Pb would limit the feasibility of using GAC to remove radon from drinking water. A detailed analysis to support and justify requirements for disposal of water treatment wastes containing naturally occurring radionuclides has not been published. The need exists for a detailed assessment of the risk associated with the disposal of GAC containing ^{210}Pb . This assessment would produce the basis for disposal requirements which would determine the feasibility of using GAC to remove radon from drinking water supplies.

INTRODUCTION

Radon-222 (^{222}Rn) is a chemically-inert radioactive gas which is an intermediate product of the decay of uranium-238 (^{238}U). It is highly mobile and is nearly ubiquitous in environmental media including the Earth's crust, air, and water. Radon-222 is a frequently encountered radiological constituent in natural waters and typically exceeds the concentration of other radionuclides including uranium, thorium, and radium by orders of magnitude (Hess et al. 1985). The wide distribution and frequently high concentrations of ^{222}Rn in drinking water supplies are of concern because a significant public health risk from cancer is associated with exposure to ^{222}Rn . Estimates of this risk show that the average lifetime probability of cancer induction from exposure to ^{222}Rn in drinking water greatly exceeds that imposed by any other natural or anthropogenic environmental contaminant in water regulated by the U.S. Environmental Protection Agency (Cothorn and Marcus 1984).

The U.S. Environmental Protection Agency has announced its intent to issue a standard for ^{222}Rn in public drinking water supplies. This standard is expected to be in the vicinity of 300 pCi/l, and many public drinking water supplies in North Carolina, as well as other states, will require ^{222}Rn removal. Also there is a need for ^{222}Rn removal systems for individual well water supplies. These individual drinking water sources, which will not be regulated by the EPA standard, have some of the highest ^{222}Rn concentrations, and removal systems are needed to reduce the risks to the persons using these sources.

RADON OCCURRENCE

The distribution of ^{222}Rn in U.S. drinking water supplies has been reported by the U.S. Environmental Protection Agency (Horton 1983; 1985) and by Hess et al. (1985). The ^{222}Rn concentration in drinking water throughout the United States ranges from near zero to over 1×10^6 pCi/l. Surface waters typically contain very little ^{222}Rn , but high concentrations may occur in groundwaters (Hess et al. 1985). The largest areas of elevated ^{222}Rn concentrations in U.S. groundwater are in New England and the Appalachian and Piedmont provinces of the Southeast. High concentrations have also been observed in groundwater in the Rocky Mountain states and California (Hess et al. 1985; Horton 1985). Groundwaters in the Atlantic-Gulf coastal plain and the midwest region have substantially lower ^{222}Rn concentrations (Hess et al. 1985; Horton 1985). In addition to variation associated with location and source, nationwide studies have shown that ^{222}Rn concentration varies among water systems of different sizes. The highest ^{222}Rn concentrations are found in private wells and small public water supplies, while large public water supplies typically have much lower ^{222}Rn content (Hess et al. 1985; Horton 1983).

Previous studies of ^{222}Rn in North Carolina groundwater have been directed toward two areas of inquiry: 1) investigating ^{222}Rn occurrence in the eastern North Carolina phosphate district, and 2) establishing the statewide distribution of ^{222}Rn in groundwater and investigating its correlates. Strain, Watson, and Fong (1979) and Watson and Mitsch (1987) studied the local effects of ore mining and processing on the ^{222}Rn content of groundwaters in the phosphate district with emphasis on geologic and hydrologic factors. These researchers did not find generally elevated ^{222}Rn concentrations in wells near mined areas and found no evidence that mining operations have induced ^{222}Rn migration into the Castle Hayne aquifer which underlies the mineralized strata.

The occurrence of ^{222}Rn in North Carolina groundwaters has been surveyed in two statewide sampling studies conducted by the Radiation Protection Section of the North Carolina Department of Human Resources (Aldrich, Sasser, and Conners 1975; Sasser and Watson 1978) and the U.S. Environmental Protection Agency (Horton 1983; 1985; U.S. EPA 1982). These investigations show that ^{222}Rn concentrations range widely in the state, from analytical zero to over 46,000 pCi/L.

Lee, Watson, and Fong (1979) investigated the relationship of ^{222}Rn and ^{226}Ra in selected North Carolina groundwaters and showed that high ^{222}Rn concentrations are generally unsupported by dissolved ^{226}Ra . Loomis (1987a, 1987b) reviewed previous statewide sampling data and investigated the relationship of groundwater ^{222}Rn concentration to bedrock geology and water system size. This work supported the earlier suggestion of Sasser and Watson (1978) that public exposure to ^{222}Rn in North Carolina may be significantly higher than for the nation as a whole. Whereas Hess et al. (1985) reported that only 15% of U.S. groundwater systems sampled contained more than 1000 pCi/l ^{222}Rn , in North Carolina 40% of 339 groundwater samples from previous statewide studies exceeded this concentration.

RADON REMOVAL

The principal methods that have been proposed and tested for removal of ^{222}Rn from water are aeration and adsorption on granular activated carbon (GAC) (Becker and Lachajczyk 1984; Kinner, Malley, and Clement 1990; Lowry 1983; Lowry and Brandow 1985; Lowry and Moreau 1986; Lowry and Lowry 1988; Lowry, Lowry, and Toppan 1988; Reid, Lassovszky, and Hathaway 1985; Rydell, Keene, and Lowry 1989). Lowry et al. have provided an excellent summary of investigations of the use of aeration and GAC for removal of ^{222}Rn from household water supplies (Lowry et al. 1987). They note that it is possible to remove ^{222}Rn at 99% efficiency with an aeration process. However, Lowry et al. point out two disadvantages of aeration processes: (1) the processes are relatively expensive, and (2) the processes are mechanical and require maintenance and additional power expenditure. On the other hand, Lowry et al. conclude "...GAC to be ideally suited to ^{222}Rn removal in point-of-entry applications." E. Roberts (Mecklenburg Purifer, Newell, N.C., pers. com. 1991) has quoted the current, installed cost of a 2 tank GAC system to be in the range of \$1,100 to \$1,400 compared to approximately \$3,500 for an aeration unit. The efficiency of ^{222}Rn removal with GAC is comparable to that with aeration. Lowry et al. state that the GAC will last for an indefinite period of time because of the small mass of ^{222}Rn and because of the short half-lives of ^{222}Rn and its first four decay products. The first four decay products of ^{222}Rn have half-lives of 3.1 minutes, 26.8 minutes, 19.7 minutes, and 1.6×10^{-4} seconds. Because these half-lives are short, an adsorption-desorption-decay steady state condition for ^{222}Rn and its first four progeny occurs within 10 to 15 days. An issue in the use of GAC is radiation exposure levels from ^{222}Rn and its progeny near the GAC during its use. This exposure can be avoided or minimized by either locating the system where it will not be a problem or by shielding the system. A second issue is the long term build-up on the GAC of ^{210}Pb , the fifth decay product of ^{222}Rn , which has a half-life of 22 years. The build-up of the radioactive ^{210}Pb could require that the GAC be disposed of as radioactive waste. Lowry et al. state that "Based on very limited data, it appears that the build-up of long-lived ^{210}Pb will not cause a disposal problem if beds are replaced every 10 to 50 years, depending on the initial ^{222}Rn concentration." However, Aieta et al. (1987) state that "The short half-life of ^{222}Rn (3.82 days) eliminates concern about hazardous waste, but the radioactive progeny ^{210}Pb may present a significant disposal problem."

PROJECT OBJECTIVES

The objectives of this project were to study the build-up of ^{210}Pb on GAC used to remove ^{222}Rn from drinking water and to investigate whether the ^{210}Pb build-up would present a problem for the disposal of the GAC. The build-up of ^{210}Pb on GAC was studied by two methods. First, calculations were performed of the activity of ^{210}Pb on GAC as a function of time of use, ^{222}Rn water concentration and water use rate. For the calculations, assumptions were made for the fraction of ^{222}Rn adsorbed onto the GAC and the fraction of ^{210}Pb retained on the GAC. Next, experimental measurements were made to determine the ^{222}Rn removal efficiency and ^{210}Pb retention fraction. An investigation was conducted of radioactive waste regulations to determine criteria for requiring that materials be disposed of as radioactive waste rather than as ordinary waste. These criteria and the results of the computations and measurements were used to evaluate whether, or under what conditions, the GAC used to remove ^{222}Rn from drinking water would be classified as radioactive waste.

DISPOSAL REQUIREMENTS

At the present time there are no federal regulations specifically applicable to the disposal of GAC containing natural radionuclides. The U.S. Environmental Protection Agency is developing standards that will include criteria for the disposal of naturally occurring radioactive material.

A survey was conducted of states with operational low-level radioactive waste disposal sites or with plans to develop disposal sites to determine whether these states have regulations applicable to the disposal of GAC containing natural radionuclides. This survey revealed that most states do not have pertinent regulations. Many states responded that they will follow guidance from the Environmental Protection Agency. The states of Illinois and Wisconsin have developed disposal criteria for water treatment plant waste containing radium. Also, the state of Washington has regulations requiring that waste containing greater than 2,000 pCi/g of natural radionuclides be disposed of as low-level radioactive waste.

In the near future the U.S. Environmental Protection Agency is expected to propose regulations controlling the disposal of waste containing naturally occurring radionuclides in excess of 2,000 pCi/g, under the authority of the Toxic Substances Control Act (U.S. EPA 1990). There is indication that at a later date EPA will consider the development of regulations addressing the disposal and use of waste that contain less than 2,000 pCi/g of naturally occurring radionuclides.

The EPA's Office of Drinking Water has recently published "suggested guidelines" for the disposal of naturally occurring radionuclides generated by drinking water treatment plants (U.S. EPA 1990). These guidelines group ^{210}Pb with radium, and for solid wastes containing 3 to 50 pCi/g (dry) of radium and ^{210}Pb the guidelines state:

These wastes should be disposed of with a physical barrier (i.e., a cover) that would protect against radon release and isolate the wastes, and provided with institutional controls designed to avoid inappropriate uses of the disposal site. A physical barrier consisting of ten feet of cover of earth or non-radioactive waste, properly designed for long-term stability of the waste, should suffice.

Sludges should be dewatered prior to disposal to minimize migration of contaminants. Consideration should be given to the hydrogeology of the site and other factors affecting long-term stability of the wastes. Sites that fully comply with EPA's Subtitle D regulations and guidance under the Resource Conservation and Recovery Act would be adequate disposal sites.

A jurisdiction may choose to ensure groundwater protection by specifying RCRA hazardous waste requirements, such as properly lined waste units, or sludge stabilization, to prevent seepage of contaminants out of the landfill. The degree of additional protection a jurisdiction wants to provide against intrusion and misuse may vary from site to site, but should be determined prior to waste disposal.

For solid wastes containing 50 to 2,000 pCi/g (dry) of radium and ^{210}Pb the guidelines state:

The disposal method should be determined case-by-case. Methods that comply with EPA's standards for disposal of uranium mill tailings should be considered (40 CFR 192).

A decision not to fully employ such methods should be based on a demonstration of significant differences between the quantity and potential for migration of uranium mill tailings versus water treatment wastes. The disposal method should be augmented by long-term institutional controls to avoid future misuse of disposal sites. Such institutional controls are not normally already in place at sanitary landfills. At a minimum, disposal in RCRA permitted hazardous waste units should be considered.

In States where NARM (Naturally-occurring and Accelerator-produced Radioactive Material) disposal is licensed or permitted, disposal at a NARM site should be considered for radium or lead-210 bearing solid wastes.

At concentrations approaching 2,000 pCi/g, disposal of wastes within a licensed low-level radioactive waste disposal facility, or a facility that is permitted by EPA or a State to dispose of discrete NARM, should be considered.

The grouping of ^{210}Pb with radium for disposal guidelines is a significant change from draft guidelines on this same subject prepared by EPA (U.S. EPA 1989). The draft guidelines grouped ^{210}Pb with uranium and for solid wastes containing 30 to 2,000 pCi/g (dry weight) uranium or ^{210}Pb stated:

The disposal method for solid wastes containing 30 to 2,000 pCi/g of uranium or lead-210 should be determined case-by-case. In general, it appears that disposal by standard municipal landfill practices would assure dilution and shielding to control external radiation. However, such disposal should be examined to ensure that sludges, in combination with other radioactive materials, do not constitute a substantial fraction (greater than about 10%) of the total wastes in the landfill. More effective physical stability and groundwater protection controls and institutional controls against misuse may be needed where large quantities of wastes are produced at the high concentration end of this range. Disposal in the manner EPA provides for uranium mill tailings and hazardous wastes should be considered.

A detailed analysis to support and justify requirements for disposal of naturally occurring radionuclides has not been published. However, it is known that radium-bearing materials are a particular concern in the disposal of naturally occurring radionuclides. The decay of ^{226}Ra produces ^{222}Rn which is an inhalation hazard. In addition, ^{226}Rn and its decay products can also present external and ingestion (from food products) hazards. Lead-210 does not have radon associated with it and would be expected to present a lesser hazard than ^{226}Ra , for equal activities. The need exists for a detailed assessment of the risk associated with the disposal of GAC containing ^{210}Pb .

COMPUTATIONS

The decay of ^{222}Rn leads to 4 radionuclides with short half-lives (^{218}Po , ^{214}Pb , ^{214}Bi , and ^{214}Po) followed by ^{210}Pb with a half-life of 22 years. When ^{222}Rn is removed from water by adsorption on GAC, the nuclides with short half-lives quickly (within approximately 4 hours) come into equilibrium with the ^{222}Rn . That is, the activity of each of these decay products will equal the activity of the ^{222}Rn on the GAC. The activity of ^{210}Pb , which has a much longer half-life, will slowly build-up on the GAC. When the GAC is removed from service, the activity of ^{222}Rn , and its short-lived progeny, will decay with a half-life of 3.8 days and after 38 days the activities of these nuclides will be decreased by a factor of approximately 10^3 . After 76 days the decrease will be approximately 10^6 . However, ^{210}Pb on the GAC will decay much slower, with a half-life of 22 years.

Since ^{210}Pb is the nuclide of concern on GAC, it is important to be able to quantify the activity of this nuclide.

During use, the rate of change with time of ^{222}Rn activity on the GAC can be described as:

$$\frac{dA_{\text{Rn}}}{dt} = CUF - \lambda_{\text{Rn}} A_{\text{Rn}}, \quad (1)$$

where A_{Rn} = ^{222}Rn activity,
 C = ^{222}Rn concentration in water,
 U = Water use rate,
 F = Fraction of ^{222}Rn adsorbed on GAC, and
 λ_{Rn} = ^{222}Rn decay constant.

The solution to this equation, assuming that $A_{\text{Rn}}=0$ at $t=0$, is:

$$A_{\text{Rn}} = \frac{CUF}{\lambda_{\text{Rn}}} (1 - e^{-\lambda_{\text{Rn}} t}) \quad (2)$$

When t becomes large (compared to the half-life of ^{222}Rn) a constant or equilibrium activity results:

$$A_{\text{Rn}} = \frac{CUF}{\lambda_{\text{Rn}}} \quad (3)$$

After 30 days, the activity of ^{222}Rn on GAC is over 99% of its equilibrium or maximum value.

The rate of change with time of the activity of ^{218}Po , the immediate decay product of ^{222}Rn , is related to the activity of ^{222}Rn (since each ^{222}Rn decay produces an atom of ^{218}Po) and, to the decay rate of ^{218}Po . This may be written:

$$\frac{dA_{Po}}{dt} = \lambda_{Po} A_{Rn} - \lambda_{Po} A_{Po}, \quad (4)$$

Where $A_{Po} = {}^{218}Po$ activity,
 $\lambda_{Po} = {}^{218}Po$ decay constant, and
 $A_{Rn} = {}^{222}Rn$ activity.

Equations for other ${}^{222}Rn$ progeny may be similarly written. However, computations of activities of the short-lived radon progeny (${}^{218}Po$, ${}^{214}Pb$, ${}^{214}Bi$, and ${}^{214}Po$) are simplified since at equilibrium (obtained in approximately 4 hours) the activity of each of these nuclides is approximately equal to the activity of ${}^{222}Rn$.

The rate of change with time of the activity of ${}^{210}Pb$ may be written:

$$\frac{dA_{Pb}}{dt} = \lambda_{Pb} A_{Po'} - \lambda_{Pb} A_{Pb}, \quad (5)$$

where $A_{Pb} = {}^{210}Pb$ activity,
 $\lambda_{Pb} = {}^{210}Pb$ decay constant, and
 $A_{Po'} = {}^{214}Po$ activity.

Since after 4 hours, the activity of ${}^{214}Po$ is equal to the activity of ${}^{222}Rn$ and after 30 days the activity of ${}^{222}Rn$ is a constant equal to CUF/λ_{Rn} ,

$$A_{Po'} = CUF/\lambda_{Rn} \quad (6)$$

for times > 30 days, and

$$\frac{dA_{Pb}}{dt} = \lambda_{Pb} \frac{CUF}{\lambda_{Rn}} - \lambda_{Pb} A_{Pb}. \quad (7)$$

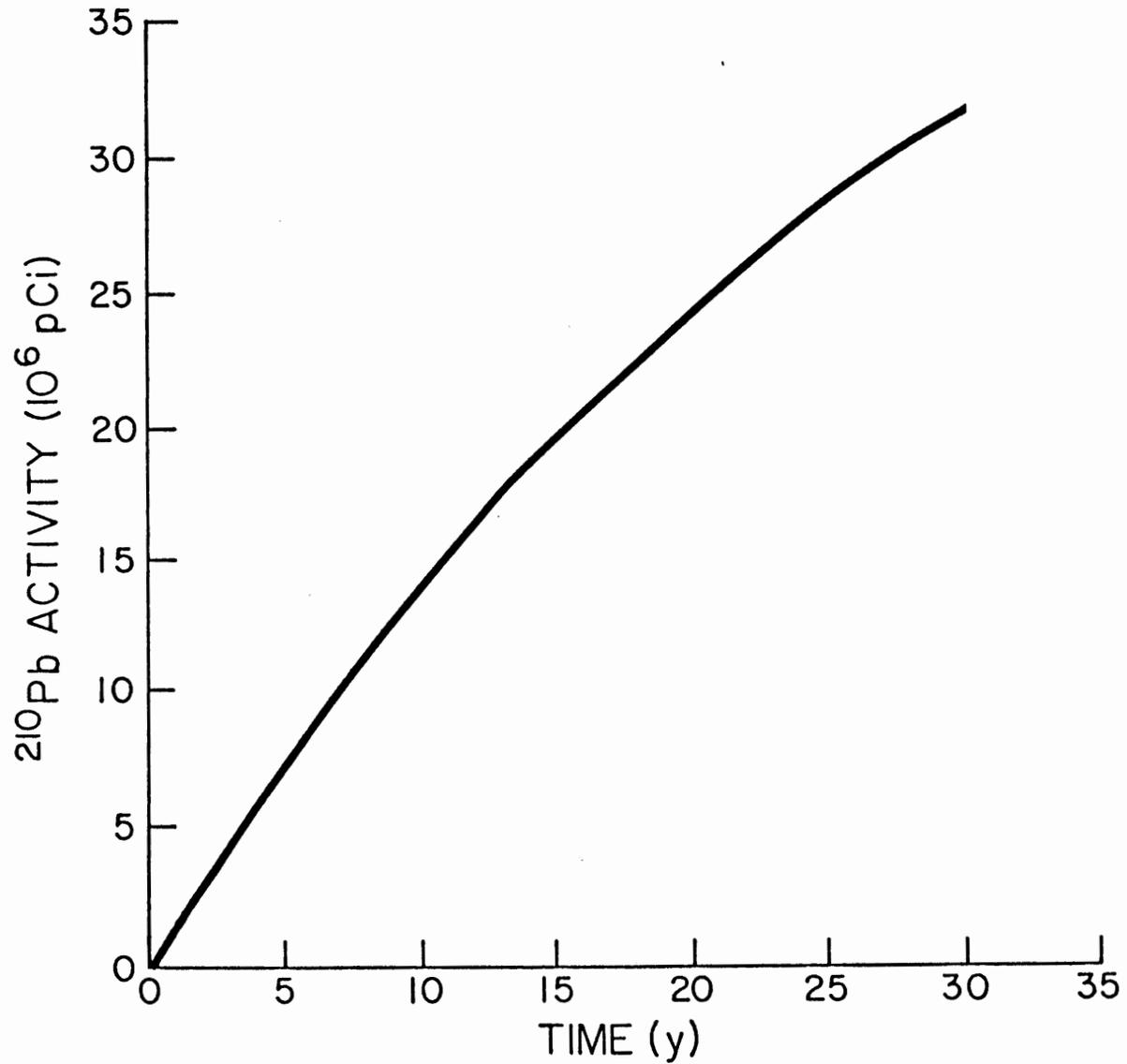
The solution to this equation is

$$A_{Pb} = \frac{CUF}{\lambda_{Rn}} (1 - e^{-\lambda_{Pb} t}). \quad (8)$$

This equation can be used to compute the activity of ${}^{210}Pb$ on GAC as a function of time when the ${}^{222}Rn$ water concentration, water use rate and ${}^{222}Rn$ removal fraction are known. This assumes 100% retention of ${}^{210}Pb$ on the GAC.

The build-up of ${}^{210}Pb$ on GAC for a ${}^{222}Rn$ water concentration of 10^4 pCi/l and a water use rate of 10^3 l/d (264 gal/d) is shown in Figure 1. This computation assumes 95% ${}^{222}Rn$

Figure 1. Build-up of ^{210}Pb on GAC for ^{222}Rn Water Concentration of 10^4 pCi/L and Water Use Rate of 10^3 L/d. Assumes 95% ^{222}Rn Removal and 100% ^{210}Pb Retention



removal and 100% ^{210}Pb retention on the GAC. (A water use rate of 10^3 l/d approximately corresponds to that used by a family of 3 persons.)

This figure can also be used to determine the ^{210}Pb activity, as a function of time, for other ^{222}Rn water concentrations and water use rates since the ^{210}Pb activity is directly proportional to the ^{222}Rn concentration and water use rate. The following equation may be used:

$$A'_{\text{Pb}}(t) = A_{\text{Pb}}(t) \frac{C'U'}{CU}, \quad (9)$$

where $A'_{\text{Pb}}(t)$ is the activity of ^{210}Pb at time t for a ^{222}Rn concentration of C' and a use rate of U' ; and $A_{\text{Pb}}(t)$ is the activity of ^{210}Pb at time t obtained from Figure 1; C is 10^4 pCi/l and U is 10^3 l/d. Thus:

$$A'_{\text{Pb}}(t) = A_{\text{Pb}}(t) \frac{C'U'}{10^7 \text{ pCi/d}}. \quad (10)$$

The concentration of ^{210}Pb on the GAC is computed by simply dividing the ^{210}Pb activity by the mass of GAC. The time required for the ^{210}Pb concentration to build-up to 2,000 pCi/l can also be computed. For calculational purposes, a 15 kg mass of GAC is assumed. The total activity of ^{210}Pb on the GAC when the concentration equals 2000 pCi/g is 3×10^7 pCi. The time, t' , at which this build-up occurs may be computed from the equation:

$$3 \times 10^7 \text{ pCi} = \frac{CUF (1 - e^{-\lambda_{\text{Pb}} t'})}{\lambda_{\text{Rn}}}. \quad (11)$$

Figure 2 shows the time at which the ^{210}Pb concentration on GAC will equal 2,000 pCi/g for ^{222}Rn water concentrations between approximately 10^4 and 10^5 pCi/l and for water use rates between 5×10^2 and 2×10^3 l/d. It is assumed that 95% of the ^{222}Rn is removed by the GAC, that 100% of the ^{210}Pb is retained on the GAC, and that the mass of GAC is 15 kg. For a ^{222}Rn water concentration of 10^4 pCi/l and a use rate of 10^3 l/d or less, over 25 years of use are required for the ^{210}Pb concentration on the GAC to exceed 2,000 pCi/l. At a use rate of 2×10^3 l/d, the concentration would not exceed 2,000 pCi/l for approximately 10 years. The majority of individual water sources in North Carolina that are likely to exceed the standard for public supplies have radon concentrations less than 10,000 pCi/l. Even for a ^{222}Rn concentration of 2×10^4 pCi/l and a use rate of 2×10^3 l/d, approximately 5 years would be required for the concentration on the GAC to reach 2,000 pCi/g. Increasing the mass of GAC would increase the time required for the concentration to reach 2,000 pCi/g.

The time required for ^{210}Pb to reach a concentration of 50 pCi/l on GAC was also computed and is shown in Figure 3. For this computation, a water use rate of 10^3 l/d, 95% ^{222}Rn removal, 100% ^{210}Pb retention and a GAC mass of 15 kg were assumed. For ^{222}Rn concentrations in water greater than 5×10^3 pCi/l, the ^{210}Pb concentration on GAC will reach 50 pCi/l in less than 1 year. The ^{210}Pb concentration on GAC will equal 50 pCi/l in approximately 5 years when the ^{222}Rn concentration in water is 1×10^3 pCi/l.

Figure 2. Time At Which ^{210}Pb Concentration on GAC Will Equal 2,000 pCi/g. Assumes 95% ^{222}Rn Removal, 100% ^{210}Pb Retention, and 15 kg GAC Mass

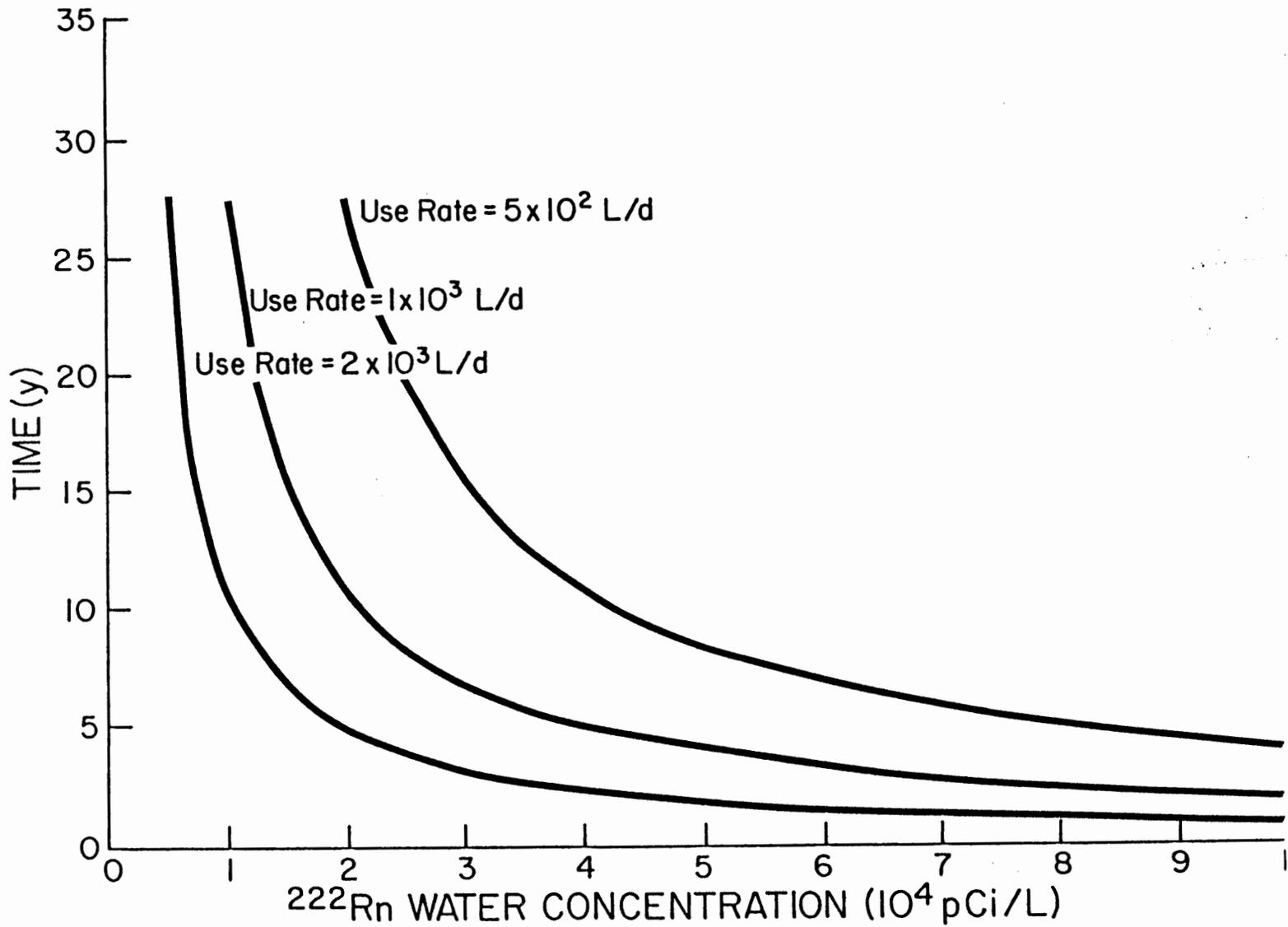
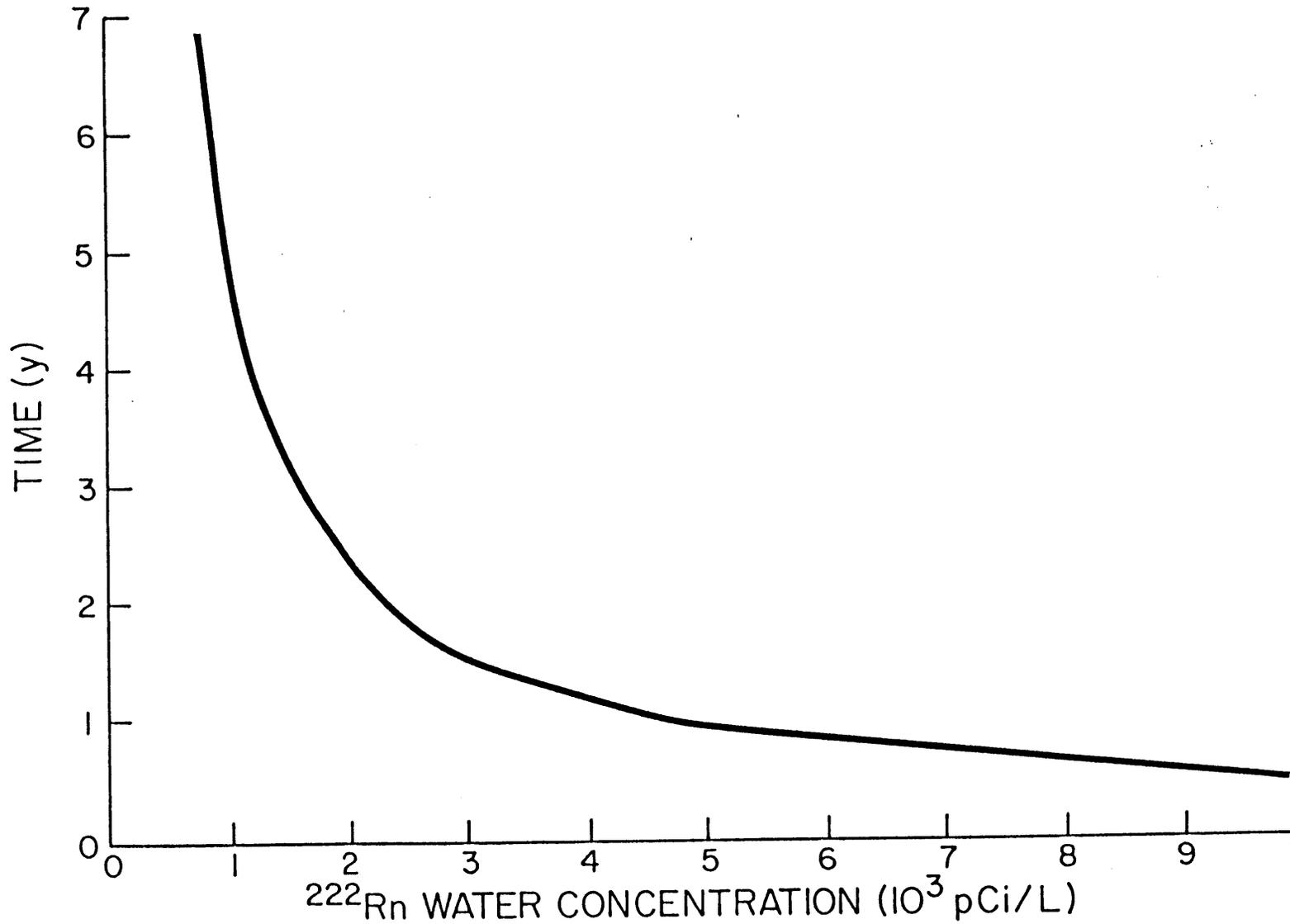


Figure 3. Time At Which ^{210}Pb Concentration on GAC Will Equal 50 pCi/g. Assumes Use of 10^3 L/d, 95% ^{222}Rn Removal, 100% ^{210}Pb Retention and 15 kg GAC Mass



EXPERIMENTAL METHODS

INTRODUCTION

The second component of this project consisted of an experimental determination of the removal fraction for radon from drinking water by granular activated carbon. In addition, the experiments were designed to test the feasibility of using photon spectroscopy of ^{210}Pb decays in quantifying retention of radon. The choice of ^{210}Pb was driven by two considerations. The first was the potential for loss of radon during transfer of the carbon from the extraction column to the counting system. The second was the finding in the review of radiological hazards that ^{210}Pb would be the primary concern in disposal.

The overall design of the experiment was to obtain a source of radon laden water of known concentration, flow the water through the extraction column, measure the inlet and outlet concentrations of radon in the water, and measure the activity of ^{210}Pb in slices of carbon from the column. While the inlet and outlet concentrations in water could be used to determine the retention fraction for radon, the outlet concentrations might in some instances be too low for reliable estimates, at least as the primary source of data. As a result, the activity of the radon decay chain (specifically the progeny ^{210}Pb) was used as the index of retention.

MATERIALS AND DESIGN

Initially, a source of radon laden water in Eastern North Carolina was identified. Use of this well proved, however, to be infeasible. The pumps associated with the well could not be adjusted to sufficiently slow flow rates to avoid rapid break through of the radon. Calculations of draw-down in the well indicated that, at the flow rate required by the well owner, there was danger of the pump being uncovered during the desired 30-day collection period. Due to accessibility of the site to traffic, it also was deemed possible that the experiment would be tampered with, invalidating results.

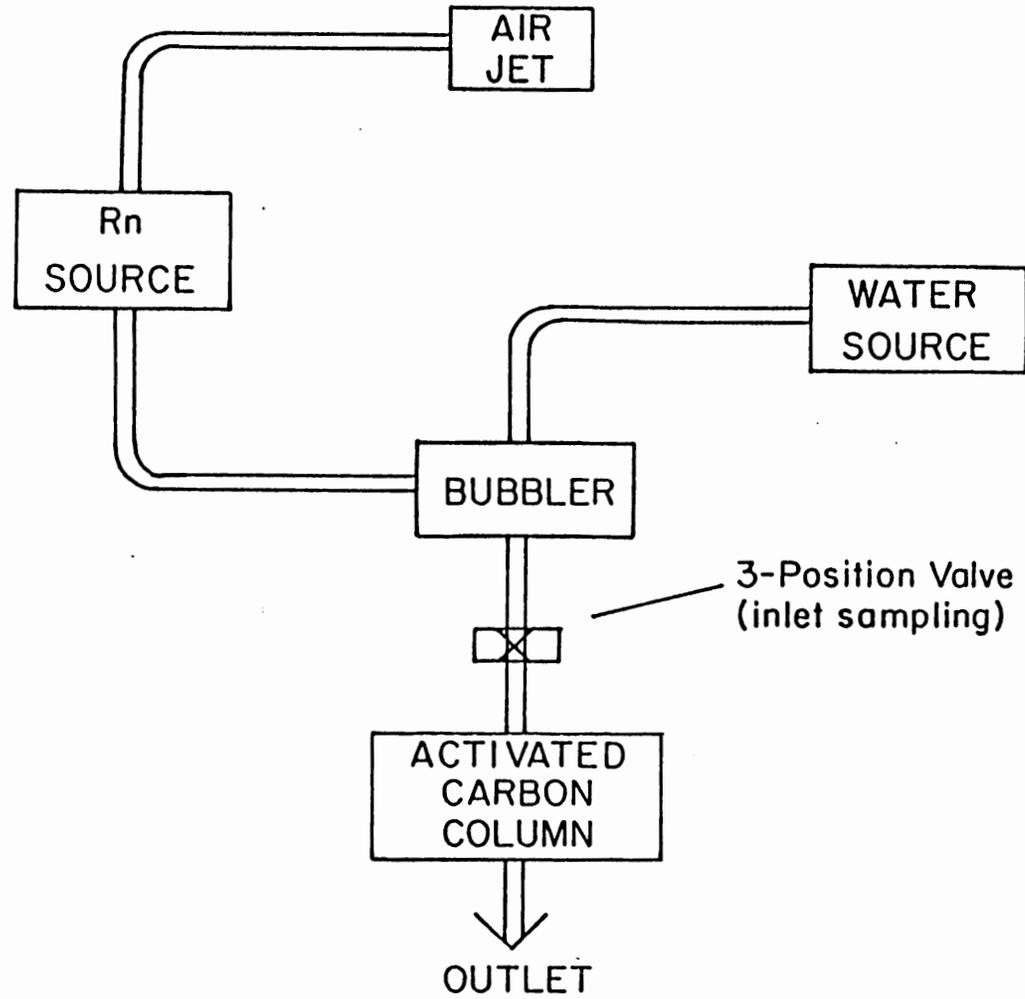
For these reasons, it was decided that a source of radon-laden water would be generated in the Radiological Laboratory of the Department of Environmental Sciences and Engineering of UNC-CH. A Pylon 1025 radon generator was employed, consisting of a 488.4 kBq source of ^{226}Ra through which air was flowed at a low rate (approximately 1 l/min). This resulted in an emission rate of 61,441 mBq of radon per minute, a rate held constant throughout the experiment.

As indicated in Figure 4, air from a constant lab bench jet was flowed through the radon generator. This flow was connected to a micro-bubbler, which allowed injection of very small bubbles (approximate diameter of 10 μ) into the flow of water. The water was obtained from a standard lab tap, flowed into the bubbler, out to a 3-position valve, and from the valve to the extraction column. The outlet of the column was placed into a drain for direct disposal.

The extraction column was a glass cylinder of length 60 cm and diameter 10.25 cm. It was packed with granular activated carbon and screens were placed at the inlet and outlet to prevent loss of carbon. A total of 22,000 grams of Calgon activated carbon was used in the experiment.

Water free of radon was flowed through the carbon for 2 days prior to the experiment to allow saturation of the column by water and to establish flow rates. The flow rate of water typically ranged between 0.5 and 1.5 l/min throughout the study period, depending on fluctuations in

Figure 4. System for Experimental Determination of Radon Removal from Water



water pressure. With the radon source connected, water was passed through the column for a total of 30 days. Inlet and outlet concentration was measured at 2 to 3 day intervals throughout this period of time, as was the flow rate of water. The flow of water was determined through collection in a tapered 2000 ml graduated cylinder and simultaneous measurement of time with a stop watch.

The concentration of radon in water for both the inlet and outlet was determined by liquid scintillation counting (LSC) of the alpha emissions from the radon decay chain at equilibrium. Water was allowed to flow into a 1000 ml graduated cylinder for approximately 5 minutes. After this time, a 30 ml sample of water was withdrawn from the bottom of the cylinder using a 30 ml syringe (water continued to flow during the withdrawal). Prior to the collection of water, three scintillation vials were prepared with 10 ml of Aquasol liquid scintillation counting cocktail. Ten ml of the collected water then was injected into each vial below the LSC cocktail (to avoid loss of the radon). Vials then were sealed and put aside for approximately 5 hours to allow ingrowth of the radon progeny and decay of any non-radon supported progeny. Samples were taken at times corresponding to the measurement of flow rate.

Analyses of the samples for alpha activity were made with a Packard liquid scintillation detection system. In all cases, samples were shaken prior to counting to ensure that the radon mixed with the cocktail. Spectral analysis first was conducted to confirm that alpha energies corresponded to those of the progeny, and to establish three regions of interest. Region A was set to channels 0 to 500, region B to channels 500 to 2000, and region C to channels 0 to 2000. Samples were analyzed until count rates in all regions were characterized by standard errors of less than 5%. The resulting count rate in each region was recorded.

Calibration was accomplished through use of an NEN radon standard available through the Physics department of UNC-CH. The standard uses a 1 Ci ^{226}Ra source to generate 10 ml samples of water (or other fluids) containing 10,000 pCi of (22,200 dpm) radon. These samples were injected by syringe into 5 samples of outlet water from the extraction column and 5 samples of inlet water, after these samples had been analyzed with the LSC. Samples then were allowed to equilibrate for 5 hours. The spiked samples (see Table 3) yielded a conversion factor of 4.1 cpm/pCi for the outlet water and 5.5 cpm/pCi for the inlet water (both efficiencies employ Region B to avoid complications of the presence of beta emitters). The higher efficiency of the inlet water presumably is due to quenching of the outlet sample by dissolved carbon.

The concentration of radon in a given sample then was obtained by dividing the count rate in Region B by the conversion factors noted above. For the inlet concentration, this yields

$$C_i = \text{CPM}_B / 5.5 V, \quad (12)$$

where V is the sample volume in liters. For the outlet concentration, this yields

$$C_o = \text{CPM}_B / 4.1 V \quad (13)$$

Both C_i and C_o then are in units of pCi/l for the radon. The total flow rate of radon into the extraction column at any point in time then equals the product of C_i and the water flow rate. The total amount of radon entering the column is the integral of the flow rate of radon (in pCi/minute) over the exposure time (30 days).

At the end of exposure, the water and radon source were disconnected from the extraction column. The column then was left to sit for approximately 14 days to allow any adsorbed radon to decay to ^{210}Pb . After this waiting period, the column was opened and divided into 9 sections of equal mass (2444 grams per section). The carbon in each section was mixed uniformly and a sample of 50 grams (dry weight) was placed into metal canisters used for radon detection. The canisters then were sealed to prevent loss of any ^{210}Pb .

Detection of the ^{210}Pb was accomplished by placing the canisters (in turn) onto a NaI detector housed in a lead shield. The detection system employed an ORTEC 113 Preamplifier (500 pf), an ORTEC 571 amplifier (gain of 175; shaping time of 6 us), a Power Design voltage supply AEC-5000 (1000V), and a TRACER NORTHERN TN-1705 multi-channel analyzer (conversion gain of 512 channels; zero offset).

Canisters were counted for approximately 30,000 seconds depending upon the ^{210}Pb activity in a sample. Detection efficiency was determined by placing a source of known activity at points uniformly distributed throughout the lowest activity slice from the column (i.e. slice #9). Cs-137 (662 KeV), ^{241}Am (60 KeV) and ^{57}Co (122 KeV) sources were used for these determinations of detection efficiency. In each case, the detection efficiencies at selected locations in the canister were obtained and averaged over the entire 50 gram mass through weighting of the detection efficiency at each location by the fraction of mass at that location. A plot of detection efficiency versus photon energy then was extrapolated to 47 KeV, the energy of the Bi L-x ray from the ^{210}Pb . This yielded a detection efficiency (including the geometry factor, attenuation and intrinsic efficiency) of 9.3% for ^{210}Pb in a 50 gram sample.

Each spectrum from the carbon slices was printed and the area under the ^{210}Pb peak determined. The location of the ^{210}Pb peak was determined using a ^{210}Pb calibration source inserted into the well of the NaI detector. Only the full energy peak was used. The precise region of interest for each spectrum was varied by a few channels in each measurement due to fluctuations in the detection system, but the Region of Interest (ROI) generally was between channels 14 and 22. Background in this region was determined by valley-to-valley averaging.

Pb-210 emits a 47 KeV x-ray in 4% of the decays. The rate of disintegration (dpm) for ^{210}Pb then is related to the total counts in the ROI by the equation:

$$\text{DPM}_{210} = \frac{(\text{CPM}_{\text{ROI}} - \text{CPM}_{\text{B}})}{0.04 \times 0.093} \quad (14)$$

where CPM_{ROI} is the count rate in the ROI, CPM_{B} is the background count rate in the ROI, 0.04 is the x-ray yield, and 0.093 is the detection efficiency.

The activity of ^{210}Pb in each sample then was multiplied by the ratio of 2444 to 50 grams to obtain the ^{210}Pb activity in each slice. These activities were summed over the 9 slices to yield the total ^{210}Pb activity on the extraction column (i.e. A_{Pb}). The total radon activity, A_{Rn} , retained by the column then is:

$$A_{\text{Rn}} = (A_{\text{Pb}}) \times (T_{\text{Pb}}) / (T_{\text{Rn}}) \quad (15)$$

where T_{Pb} is the half-life of ^{210}Pb (22 years) and T_{Rn} is the half-life of ^{222}Rn (3.83 days or 0.0105 years). This yields:

$$A_{\text{Rn}} = A_{\text{Pb}} \times 2095 \quad (16)$$

RESULTS

The mean flow rate of water throughout the 30 day exposure period was 0.73 l/min, yielding a total flow of 31,536 l. The mean inlet concentration of radon in the water was 2093 pCi/l, yielding a total inlet of 6.60×10^7 pCi. In a previous section, it was noted that the emanation rate from the source was 61,441 mBq per minute or 7.17×10^7 pCi in 30 days, indicating a loss of 8% of the radon between the source and the inlet. Since the system was located within, and adjacent to a hood, this radon presumably was carried to the outside air.

As will be noted from Table 1, essentially all of the activity of ^{210}Pb was contained on the first 3 slices, or the first third of the column. The activities in these slices were summed to obtain the total ^{210}Pb on the extraction column. The area under the ^{210}Pb peak in slices 4 through 9 was not sufficiently elevated to allow accurate determination of activity in these slices.

The results of the water flow rate measurements, inlet radon concentration and radon outlet concentration, are given in Table 2. The count rates in Region B of the LSC are presented in Table 3.

The total estimated activity of ^{210}Pb on the extraction column was 3.03×10^4 pCi. Using equation 16, this indicates a total radon activity of

$$A_{\text{Rn}} = 3.03 \times 10^4 \times 2095 = 6.34 \times 10^7 \text{ pCi.}$$

The calculated retention efficiency for radon is:

$$\begin{aligned} \text{Efficiency} &= 6.34 \times 10^7 \text{ pCi} / 6.60 \times 10^7 \text{ pCi} \\ &= 0.961, \end{aligned}$$

or approximately a 96% retention efficiency averaged over the 30 days of operation.

A similar calculation can be performed using the ratio of the LSC results for the inlet and outlet concentrations. The mean concentration of radon in the inlet water was 2093 pCi/l. The mean concentration of radon in the outlet water was 92.3 pCi/l. This indicates a retention efficiency for radon of:

$$\text{Efficiency} = 1 - 92.3/2093 = 0.955$$

or approximately 96% (the same as determined from the activity on the column).

The results of this study confirm the work reported by others (Lowry and Lowry 1988). The retention efficiency determined in this study is the average over a 30-day period of operation. The efficiency may decrease for longer periods of operation (Kinner, Malley, and Clement 1990).

In addition to the above measurements, the LSC samples from sample date 9-9-90 were remeasured at 4 day intervals. The activity in the sample decreased with a half-time of approximately 3.6 days. This half-time is slightly less than that of radon (3.83 days), and may reflect slight loss due to emanation of radon from the LSC vials. The data indicate that radon was, however, the source of radioactivity measured within the outlet and inlet samples. The results of these repeated measurements are provided in Table 4.

Table 1. Measurement Results for NaI Analysis. Total Counting Time was 60,000 Seconds

Slice	ROI*	CPM in ROI (w/Bkg)	CPM in ROI (w/o Bkg)	²¹⁰ Pb Activity (pCi)
1	14-21	8.5	1.3	7,550
2	14-21	9.6	3.0	17,516
3	13-20	7.9	0.9	5,134
4	14-21	8.7	ND	ND
5	13-20	10.3	ND	ND
6	15-22	9.2	ND	ND
7	14-21	9.4	ND	ND
8	14-21	8.4	ND	ND
9	14-21	8.8	ND	ND

* Region of Interest or Channel Numbers for ²¹⁰Pb peak
 CPM = Counts Per Minute
 ND = Not Detectable

Table 2. Water Flow Rate and Radon Concentration Measurements

Date	Flow Rate (l/min)	Inlet Radon (pCi/l)	Outlet Radon (pCi/l)
9-1-90	0.54	1984.9	123.1
9-4-90	0.65	2218.4	100.6
9-7-90	1.02	2295.6	74.5
9-9-90	0.82	2091.6	82.5
9-12-90	0.59	1846.9	115.5
9-15-90	0.65	2319.5	92.6
9-17-90	0.70	2006.1	84.9
9-20-90	0.86	2095.2	103.5
9-22-90	0.91	1984.6	95.2
9-26-90	0.66	1842.7	94.8
9-29-90	0.72	2201.4	70.6
9-30-90	0.64	2231.5	69.8
MEAN	0.73	2093.2	92.3

Table 3. Raw Data Obtained for the Inlet and Outlet Water Measurements

Sample Date	Inlet CPM in Region B	Outlet CPM in Region B
9-1-90	109	5.0
9-4-90	122	4.1
9-7-90	126	3.1
9-9-90	115	3.4
9-12-90	102	4.7
9-15-90	128	3.8
9-17-90	110	3.5
9-20-90	115	4.2
9-22-90	109	3.9
9-26-90	101	3.9
9-29-90	121	2.9
9-30-90	123	2.9
SPIKED*	54757.3	41382.9

* Using spike of 10,000 pCi for samples on 9-15-90. The conversion factor for the inlet then is $(54757.3-128)/10^4=5.5$ CPM/pCi and for the outlet $(41382.9-3.8)/10^4=4.1$ CPM/pCi.

CPM = Counts Per Minute
 Region B = Channels 500 to 2,000

Table 4. Half-time Determination for Inlet Water Sample Obtained on 9-9-90.

Date of Measurement	CPM in Region B
9-9-90	11503.8
9-21-90	2700.4
9-25-90	1321.5

CPM = Counts Per Minute
 Region B = Channels 500 to 2,000

REFERENCES

- Aieta, E.M., J.E. Singley, A.R. Trussell, K.W. Thorbjarnarson, and M.J. McGuire. 1987. Radionuclides in drinking water: an overview. American Water Works Association Journal. 79:144-152.
- Aldrich, L.K., M.K. Sasser, and D.A. Conners. 1975. Evaluation of radon concentrations in North Carolina ground water supplies. North Carolina Department of Human Resources, Raleigh, NC.
- Becker, A.P. III, and T.M. Lachajczyk. 1984. Evaluation of waterborne radon impact on indoor air quality and assessment of control options. U.S. Environmental Protection Agency, EPA-600/7-84-093.
- Cothem, C.R., and W.L. Marcus. 1984. Estimating risk for carcinogenic environmental contaminants and its impact on regulatory decision making. Regulatory Toxicology and Pharmacology. 4:265-274.
- Hess, C.T., J. Michel, T.R. Horton, H.M. Prichard, and W.A. Coniglio. 1985. The occurrence of radioactivity in public water supplies in the United States. Health Physics 48:553-586.
- Horton, T.R. 1983. Methods and results of EPA's study of radon in drinking water. U.S. Environmental Protection Agency, EPA. 520/5-83-027.
- Horton, T.R. 1985. Nationwide occurrence of radon and other natural radioactivity in public water supplies. US Environmental Protection Agency, EPA-520/5-85-008.
- Kinner, N.E., J.P. Malley, Jr., and J.A. Clement. 1990. Radon Removal Using Point-of-Entry Water Treatment Techniques. U.S. Environmental Protection Agency, EPA/600/S2-90/047.
- Lee, R.D., J.E. Watson, and S.W. Fong. 1979. An assessment of radium in selected North Carolina drinking water supplies. Health Physics. 37:777-779.
- Loomis, D.P. 1987a. Radon-222 concentration and aquifer lithology in North Carolina. Ground Water Monitoring Review. 7:33-39.
- Loomis, D.P. 1987b. The relationship between water system size and ²²²Rn concentration in North Carolina public water supplies. Health Physics. 52:69-71.
- Lowry, J.D. 1983. Removal of radon from water using granular activated carbon adsorption technology. Transfer Report, Land and Water Resources Center, University of Maine and the Office of Water Resources, Department of the Interior, Washington, DC.
- Lowry, J.D., and J.E. Brandow. 1985. Removal of radon from water supplies. Journal of Environmental Engineering Division - ASCE. Vol. III, No. 4.
- Lowry, J.D., and E. Moreau. 1986. Removal of extreme radon and uranium from a water supply. Proceedings of 1986 ASCE National Conference on Environmental Engineering, Cincinnati, Ohio.

- Lowry, J.D., W.F. Brutsaert, T. McEnerney, and C. Molk. 1987. Point-of-entry removal of radon from drinking water. American Water Works Association Journal. 79:162-169.
- Lowry, J.D., and S.B. Lowry. 1988. Radionuclides in Drinking Water. American Water Works Association Journal. 80:51-64.
- Lowry, J.D., S.B. Lowry, and W.C. Toppan. 1988. New Developments and Considerations for Radon Removal from Water Supplies. Proceedings of U.S. Environmental Protection Agency Symposium on Radon and Radon Reduction Technology, Denver, CO.
- Reid, G.W., P. Lassovszky, and S. Hathaway. 1985. Treatment, waste management and cost for removal of radioactivity from drinking water. Health Physics. 48:671-694.
- Rydell, S., B. Keene, and J.D. Lowry. 1989. Granulated Activated Carbon Water Treatment and Potential Radiation Hazards. Journal of New England Water Works Association. December 1989: 234-248.
- Sasser, M.K, and J.E. Watson. 1978. An evaluation of the radon concentration in North Carolina ground water supplies. Health Physics. 36:667-671.
- Strain, C.D., J.E. Watson, and S.W. Fong. 1979. An evaluation of radium-226 and radon-222 concentrations in ground and surface water near a phosphate mining and manufacturing facility. Health Physics. 37:779-783.
- U.S. Environmental Protection Agency. 1982. Environmental Radiation Data Report 28, EPA 520/1-83-002.
- U.S. Environmental Protection Agency. 1989. Suggested guidelines for the disposal of naturally occurring radionuclides generated by drinking water treatment plants. (Draft) Office of Drinking Water, Criteria and Standards Division, Washington, DC.
- U.S. Environmental Protection Agency. 1990. Suggested guidelines for the disposal of naturally occurring radionuclides generated by drinking water treatment plants. Office of Drinking Water, Criteria and Standards Division, Washington, DC.
- Watson, J.E., and B.F. Mitsch. 1987. Groundwater concentrations of ^{226}Ra and ^{222}Rn in North Carolina phosphate lands. Health Physics. 52:361-365.