United States Environmental Protection Agency

Risk Reduction Engineering Laboratory Cincinnati, OH 45268

Research and Development

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Project Summary

Radon Removal Techniques for Small Community Public Water Supplies

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The report summarized here presents the results of an evaluation of radon removal in small community water supplies with the use of full-scale granular activated carbon adsorption, diffused bubble aeration, and packed tower aeration. Various low technology alternatives, such as loss in a distribution system and addition of coarse bubble aeration to a pilot-scale atmospheric storage tank, were also evaluated.* The full report discusses each of the treatment alternatives with respect to their radon removal efficiency, potential problems (i.e., waste disposal, radiation exposure, and intermedia pollution), and economics in small community applications. In addition, several sampling methods, storage times, scintillation cocktails, and extraction procedures currently used in the liquid scintillation technique for analysis of radon in water were compared.

This Project Summary was developed by EPA's Risk Reduction Engineering Laboratory, Cincinnati, OH, to announce key findings of the research project that is fully documented in a separate report of the same title (see Project Report ordering information at back).

Introduction

As part of the 1986 amendments to the Safe Drinking Water Act, the U.S. Environmental Protection Agency (EPA) is to propose a rule for Maximum Contaminant Level Goals (MCLGs) and National Primary Drinking Water Regulations (NPDWR) including Maximum Contaminant Levels (MCLs) for radionuclides in drinking water. One of the radionuclides that will be regulated under the rule is radon-222 (radon). EPA is considering setting the MCL for radon in the range of 200 to 2,000 pCi/L. Data on the distribution of radon in groundwater supplies in the United States indicate that a large number of individual and public water supplies will be affected by an MCL in that range. In addition, many of these public water supplies will be ones serving small communities (< 76 m^3/day).

The rule will also contain recommendations with respect to Best Available Technologies (BATs) and analytical methods. Three conventional water treatment technologies (granular activated carbon (GAC), diffused bubble aeration, and packed tower aeration) have been used to remove radon from drinking water. The GAC process, which has been used to treat point-of-entry and small community water supplies, relies on the ability of radon to adsorb to the carbon. One unique aspect of this process is that the breakthrough/exhaustion profile typically seen when GAC is treating conservative (nondecaying) contaminants is not exhibited during radon removal.

The aeration methods are used because radon is a highly volatile gas with a relatively large Henry's constant (2.80 atm • $m^3 H_2O/m^3$ air at 100°C) that can be easily transferred from water to air. Aeration methods have been used in

* Environmental Research Brief EPA/600/M-87/031 also outlines the results of the low technology study.

individual, small community, and other public water supplies.

The primary purpose of this study was to evaluate the performance of full-scale GAC and diffused bubble and packed tower aeration systems when treating small community water supplies containing radon. In addition, several low technology alternatives and various modifications of the liquid scintillation counting technique used for analysis of radon in water were evaluated.

The specific objectives of the study were to:

- Evaluate full-scale GAC systems operating at two small communities in New Hampshire, by monitoring them for changes in radon removal, radiation emissions, and general water quality parameters (e.g., pH, iron, turbidity, microbial numbers);
- Conduct several specific monitoring events of the GAC systems to assess the effect on GAC performance of diurnal variations in water flowrate and raw water quality, high water flowrate, and backwashing;
- Core the GAC after several months of operation to determine if iron, manganese, microorganisms, radionuclides, or all of them were accumulating in the units;
- Evaluate full-scale diffused bubble and packed tower aeration systems operating in small communities in New Hampshire by monitoring them for radon removal, general water quality parameters, and off-gas emissions of radon;
- Operate the aeration systems over a range of volumetric air to water (A:W) ratios at two water flowrates to determine the effect of these parameters on radon removal;
- Evaluate three randomly packed plastic media in the tower aeration system with respect to radon removal efficiency;
- Evaluate the radon removal efficiency of several low technology modifications (e.g., free fall vs. bottom entry, spray nozzle entry, venturi entry, coarse bubble aeration) retrofitted to an existing small community atmospheric storage tank;
- Assess the effect of sampling techniques (e.g., free fall, hose connector, direct syringe collection, volatile organic analysis (VOA) bottle collection), storage time, scintillation cocktail, and extraction via shaking on the liquid scintillation analytical technique for analysis of radon in water.

Analytical Methods

Standard Methods and EPA methods were used to determine radon; gamma/beta emissions; the activity of total uranium, radium-226, and lead-210; microbial counts; pH; temperature; dissolved oxygen; turbidity; total iron; and manganese. The University of New Hampshire and the State of New Hampshire conducted the analyses; when commercial equipment was used, it was calibrated according to the manufacturer's directions.

Granular Activated Carbon

Downflow GAC systems were installed at two mobile home parks located in Amherst and Mont Vernon, NH. The Amherst water system serves 56 homes at an average daily flow of 59 ±4 m^3/day. The water is obtained from one well containing an average radon activity of 49,500 ±11,200 pCi/L. The Mont Vernon system supplies 40 homes at an average flow of 31 ±11 m^3/day. Water is obtained from two wells with an average radon activity of 222,000 ± 52,000 pCi/L. The well water in both systems was pumped to unpressurized (atmospheric) storage tanks and, subsequently, pumped through the GAC systems upon demand from the community.

The systems were evaluated according to the study objectives. Radon and several general water quality parameters (alkalinity, turbidity, dissolved oxygen, temperature, pH, iron, manganese, and bacterial numbers) were monitored at each site. Uranium and radium were also monitored in the water supply. The GAC was cored and analyzed for accumulation of uranium-238, uranium-235, radium-226, lead-210, iron, manganese, and microbial numbers. Gamma radiation measurements were taken at the surface of the units and at locations inside and outside of the pumphouses to determine whether exposure presented significant health and safety problems. The effects of diurnal variation in loading and backwashing were also evaluated.

The GAC used in both systems was Barneby Cheney Type 1002.* At Amherst, the system consisted of one 91.4-cm-diameter fiberglass tank containing 0.85 m^3 of GAC. At Mont Vernon, the system consisted of two contactors in series: a 76.2-cm-diameter contactor (GAC #1) containing 0.57 m^3 of GAC followed by a 91.4-cm-diameter contactor (GAC #2) containing 0.76 m^3 of GAC. The system at Mont Vernon was designed with two units because of the high influent radon activity. Taps for collecting water samples were installed on the influent and effluent lines and at 10 intervals in each GAC system.

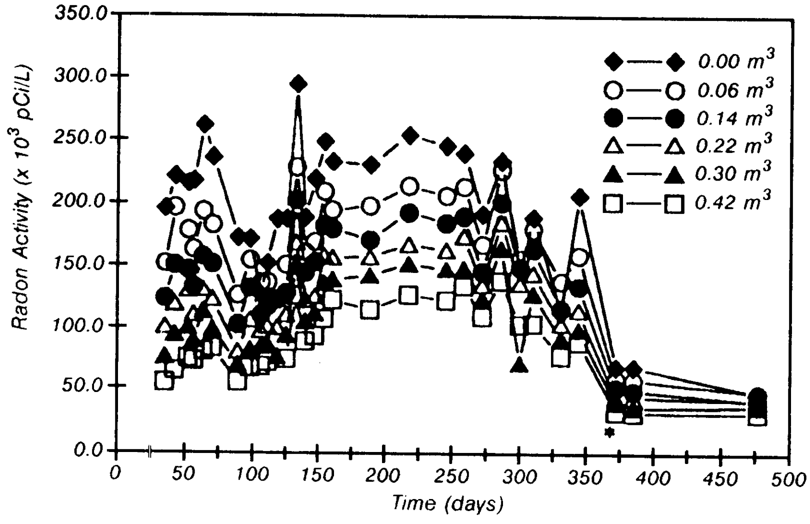
The GAC systems were monitored daily for 3 to 4 days and then every 2 to 5 days for approximately 1 month during initial operation (Phase I). Thereafter, during Phase II, they were monitored weekly, biweekly, and then monthly. The system at Amherst was monitored for 122 days, and the system at Mont Vernon was monitored for 478 days.

During the first few days of operation at Mont Vernon, all of the radon present in the water was removed in GAC #1. The radon removal front moved through the units over time, eventually breaking through into the effluent by Day 25. A similar pattern was observed with the Amherst system.

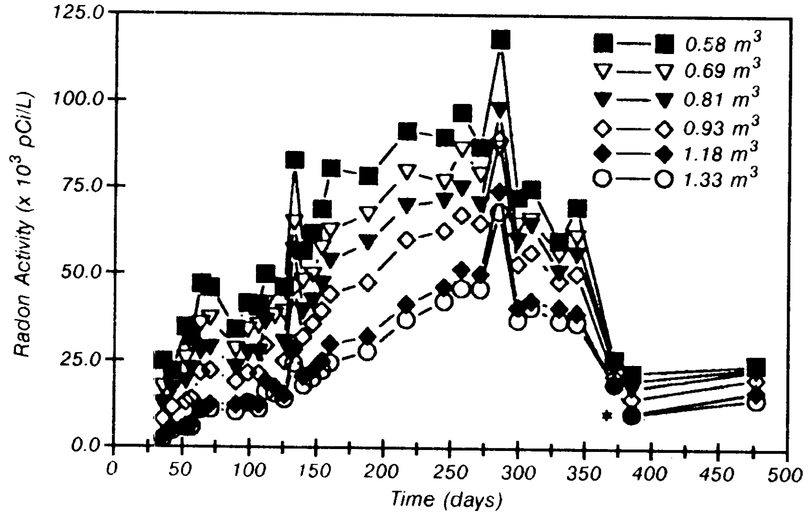
The average influent radon activity (210,491 ± 41,384 pCi/L) at Mont Vernon remained higher than the design influent activity of 155,000 pCi/L. The effluent quality during this period varied from 4,750 to 68,400 pCi/L (Figure 1). In addition, the GAC system at Mont Vernon had a higher average water flowrate $\bar{x}$=36 ± 12 m^3/day) than it was designed to handle (25 m^3/day). As a result, the overall radon loading applied to the system was usually higher than anticipated, which may have accounted for some of the increase in effluent radon activity. Radon removal followed an exponential pattern as a function of bed volume (Figure 2). Similar results were obtained at the Amherst site. During the final 3.5 months of the study at Mont Vernon, when a new well was operating (influent radon activity = 68,900 ± 1,400 pCi/L), the data showed a steep (almost straight) pattern and an overall decrease in removal efficiency (e.g., Day 477). Although this decrease in removal efficiency corresponded to changes in other water quality parameters, the effects of raw water quality on radon adsorption by GAC are poorly understood, and predictions about removal are difficult to make.

The gamma/beta emissions data obtained at both sites were in the 10^0 to 10^1 mR/hr range at the units' surfaces. This range is considerably greater than the background values of 0.03 to 0.06 mR/hr and the National Council on Radiation Protection guidelines of an 8-hr maximum exposure in residences of

* Mention of trade names or commercial products does not constitute endorsement or recommendation for use



a. GAC #1 Contactor



b. GAC #2 Contactor

Figure 1. *Phase II GAC System – Mont Vernon, NH. Radon activity through GAC #1(a) and #2(b) through 477 days operation. Note scale difference between a and b. (* New well began operating.)*

0.058 mR/hr. Though exposures are highly dependent on distance from the source, they would need to be minimized to meet accepted occupational safety and health standards, perhaps by using shielding.

Several potential problems were observed with the GAC, including accumulation of iron, uranium-238 and -235, radium-226, and lead-210, and release of bacteria. The GAC provides a good surface for bacterial attachment and concentrates the nutrients needed by microorganisms. At Mont Vernon, the effluent contained as many 10^4 CFU/mL, probably, in part, because of high influent numbers; at Amherst the effluent ranged from 200 to 400 CFU/mL. Coring data indicated that iron precipitates were retained in the GAC units. This occurred even when there was little change in the influent iron concentration as compared with that of the effluent because of the large volume of water passing through the units over time.

Uranium profiles obtained from Mont Vernon during Phase II and from core samples indicated that uranium was removed exponentially through the GAC system. This contrasted with the lack of uranium removal observed at Amherst. The discrepancies may be explained by the difference in pH of the raw waters at the two sites (Amherst = 8.03 ± 0.14; Mont Vernon = 6.5 ± 0.2). The predominant uranium species between pH 7 and 8 are soluble anionic carbonate complexes in natural waters, whereas at pH <6.8, the neutral UO_2CO_3 species predominates. It is hypothesized that the poorly adsorbed anionic species predominated at Amherst, whereas the favorably adsorbed neutral species predominated at Mont Vernon.

At both sites, radium-226 was accumulated, as determined by coring. Since radium adsorption to GAC is considered unfavorable, it is hypothesized that radium was removed by adsorption or ion exchange reactions occurring with either solid phases (e.g., $Fe(OH_3)$, $Mg(CO_3)$) or

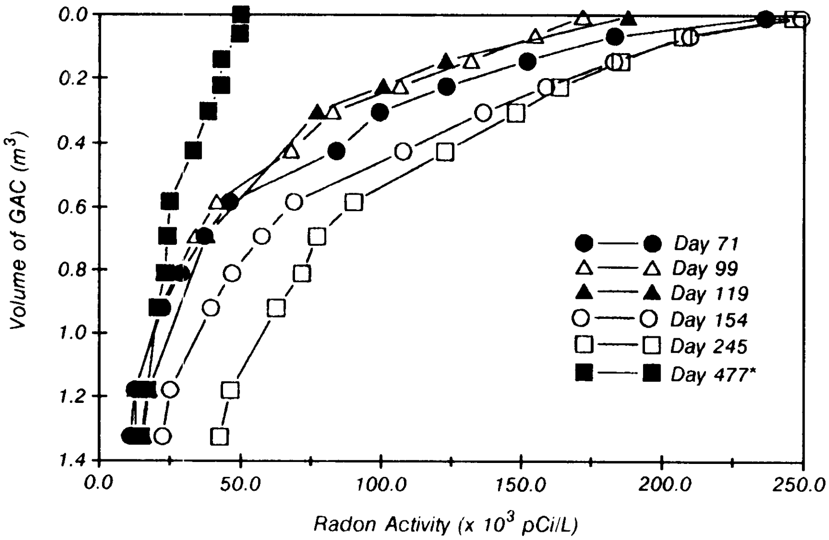


Figure 2. *Phase II GAC System – Mont Vernon, NH. Representative profiles of radon activity through the GAC system. (* New well began operating.)*

organic matter deposited in the GAC. A comparison of the theoretical lead-210 adsorbed (as a function of radon removal) and the lead-210 coring results indicated that most, if not all, of the radon progeny were retained by the GAC. Lead-210 accumulations on the GAC at the sites ranged from 10^5 to 10^6 pCi/kg, with the total beds containing between 10^7 to 10^8 pCi. The GAC used at Mont Vernon and Amherst exceeded the State of New Hampshire Radiological Health Program *de minimus* regulations for uranium-238 (58,410 pCi/kg; 2.5 x 10^{-5} Ci/m^3) and radium-226 (44.39 pCi/kg; 1.4 x 10^{-8} Ci/m^3). There are no regulations in New Hampshire for lead-210. As a result, all of the GAC used in this study was classified as a low level radioactive waste. It should be noted that the State of New Hampshire has stringent *de minimus* standards with respect to uranium and radium, and that in many other states the material would not be considered a low level radioactive waste. However, other states may regulate lead-210, which was present at much higher activities.

A detailed economic evaluation, including only costs related to installation and operation of the GAC systems, was performed. Direct capital costs were determined from expenditures made during the project, and indirect capital costs were calculated based on fixed percentages developed by the EPA Office of Drinking Water. Annual costs were developed by adding the amortized (9% interest over 20 yr) total capital cost to the annual operation and maintenance cost. Production costs were calculated by dividing the annual cost by the annual design flow. All cost figures were updated to second quarter 1989 dollars (ENR = 426). Costs are presented for comparative purposes only, since actual system costs will vary and be site specific.

The production costs for the GAC systems, including pretreatment for iron and manganese and disposal of the spent GAC as a regulated low-level radioactive waste, were estimated to be $2.15/1,000 gal for Amherst and $2.64/1,000 gal for Mont Vernon.

Diffused Bubble Aeration

A diffused bubble aeration system, consisting of three polyethylene tanks aligned in series (holding capacity of each = 1022L), was installed in a small community public water supply in Derry, NH. An air blower with a 2.66 m^3/min capacity that forced outdoor air into diffusers provided aeration. The diffusers consisted of 1.90-cm-diameter coiled plastic tubes with 0.038-cm holes drilled in their underside (spacing between holes = 0.5 to 1.6 cm). The diffusers were located 79 cm below the water's surface and 36 cm above the bottom of each tank. The radon stripped from the water was vented outside the building housing the units.

The system allowed removal efficiencies to be compared over a wide range of influent radon activities (Tank 1: 60,843 to 86,355 pCi/L radon; Tank 2: 10,096 to 80,271 pCi/L; and Tank 3: 1,767 to 74,112 pCi/L). Two flow ranges were obtained by manually operating one (low flow = 0.047 ± 0.00053 m^3 H_2O/min) or two wells (high flow = 0.10 ± 0.0019 m^3/min). Radon activities averaged 65,487 ± 5,657 pCi/L during high water flow and 78,385 ± 6,120 pCi/L during low water flow. The two water flowrates and the two radon activities resulted in applied radon loading rates averaging 6,819 ± 548 nCi (10^3 pCi) per minute (nCi/min) for high water flow and 3,639 ± 295 nCi/min for low water flow. A:W ratios of 2:1, 3:1, 5:1, 7:1, 10.5:1, 15:1, and 20:1 were tested for both water flowrates. The tanks were drained at the end of each run and refilled with raw water immediately before the start of a new run.

Figure 3 shows the overall percent removals of radon loading versus A:W ratios for the high and low water flowrates. These data were obtained after steady state conditions were achieved in the diffused bubble system. As the A:W ratio increased from 2:1 to 5:1 for each flowrate, there was a sharp increase in radon removal. Above 5:1, however, there was much less improvement in efficiency with large increases in A:W ratio.

When operating at A:W ratios of 5:1 and greater (at both high and low water flowrates), the overall radon removal efficiency ranged from 90.0% to >99.6%; the greatest efficiency was obtained at A:W ratios of 15:1 and 20:1. At A:W ratios of 10.5:1 and greater for the low flowrate and 15:1 and greater for the

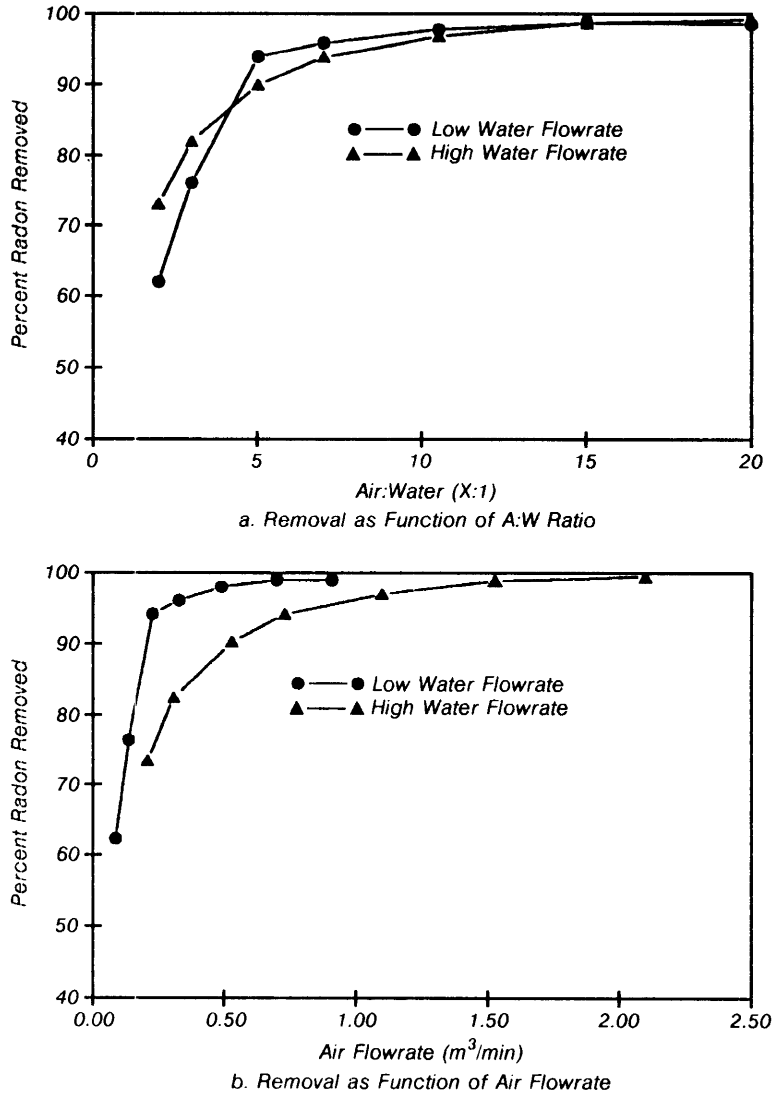


Figure 3. *Diffused bubble aeration – Derry, NH. Percent radon removal as a function of (a) A:W ratios and (b) air flowrate for the low and high water flowrate.*

high flowrate, there was no significant difference in removal efficiency (α = 0.05 and 0.01, Analysis of Variance (ANOVA)). Hence, for the diffused bubble system tested at the given conditions of radon loading, the lowest A:W ratios to yield (statistically significant) maximum radon removal for low and high water flowrates were in the range 7:1 to 10.5:1 and 10.5:1 to 15:1, respectively. From the practical viewpoint of designing a diffused bubble system, the A:W ratio should also be based on the most cost effective blower size and mode of operation.

An evaluation of individual tank performance indicated that radon removal in the diffused bubble system was a function of mass transfer. As the radon activity became progressively lower through the series of tanks, the driving force (Radon $\text{Activity}_{\text{Water}}$ - Radon $\text{Activity}_{\text{Air}}$) decreased, limiting removal. Though mass transfer may make it more difficult to achieve low effluent activities, the diffused bubble system tested produced water with radon activities of 1,849 to 280 pCi/L for A:W ratios $\geq$10.5:1. These data suggest that it may be possible to meet an MCL of 200 pCi/L, if air flowrate is high, or there is a long contact time, or both.

Stack emissions were monitored to determine if the off-gas radon activities from the diffused bubble system could affect the air quality in the surrounding environment. For the system tested, the off-gas activities (3,361 to 18,356 pCi/L) would need to be diluted 10^4 to 10^5 times to be similar to radon activities found in the ambient air at the site (0.1 to 0.15 pCi/L).

An economic evaluation, similar to that done for the GAC systems, was performed for the diffused bubble system. The total production cost, including pretreatment for iron and manganese and assuming no required treatment of the off-gas, was estimated to be $2.14/1000 gal.

Packed Tower Aeration

The packed tower aeration system was installed at the same mobile home park in Mont Vernon, NH, used in the GAC study. The system consisted of a 5.49-m-tall, 0.30-m-diameter stainless steel tower containing randomly packed plastic media. Raw water was pumped to the top of the tower and distributed by a nozzle located 15.2 cm above the top of the media. Air entered the tower 0.15 m below the media.

The major focus of the study was a series of separate 3-hr runs designed to determine the tower's radon removal efficiency for a variety of operating conditions. Parameters varied included packing type, packing height, liquid loading rate, and volumetric A:W flow ratios. Though fluctuations in water flowrate and radon activity at the site during the study prevented comparisons of the effects of A:W ratio and packing types, there was relatively little difference in the overall percent radon removal observed (92.7% to 99.8%) among the conditions tested. This was surprising considering the variation in water flowrate (0.18 to 2.6 m^3/hr), influent radon activity (115,225 to 278,488 pCi/L), and packing type. The resilience of the tower system is encouraging, considering that many small communities may experience variations in water flowrate and radon activity similar to those observed at the Mont Vernon site. It is hypothesized that the consistently higher removals occurred because radon is a highly volatile gas and because the packing height used (~3.7 m) was great enough to compensate for large variations in loading.

Most of the radon removal (Figure 4) occurred in the top 0.3 m of the tower, probably because of end effects of free fall, liquid distribution, and turbulence. The data indicate that mass transfer limitations may be a major factor in designing towers to achieve very low radon activities. Therefore, to meet an MCL of 200 pCi/L with water supplies containing moderate to high radon activities, towers may be impractical because of the extremely large packing heights required.

As observed with the diffused bubble system, radon activities in the stack emissions from the tower were extremely high and required dilutions of 10^4 to 10^5 to approach the ambient air activities (0.1 to 0.15 pCi/L) at the site.

An economic evaluation, similar to that done for the GAC systems, was performed for the packed tower. The total production cost, including pretreatment for iron and manganese and assuming no required off-gas treatment, is estimated to be $2.10/1,000 gal.

Liquid Scintillation Technique

The effects of sampling technique (direct collection using a syringe versus filling a VOA bottle), storage (up to 21 days), and choice of scintillation cocktail (toluene- and mineral-oil-based cocktails and Opti-Fluor 0) on the liquid scintillation (LSC) analytical technique for radon in water were evaluated. Other experiments were conducted to determine the sources of variability in the method (field, preparation, instrument) and the validity of the extraction via shaking procedure currently defined by EPA.

Numerically, the direct syringe sampling technique always yielded the highest radon activities, whereas the VOA bottle filled with free falling water yielded the lowest. This is not surprising because less sample handling before injection into the scintillation vial and less agitation during sampling should result in less loss of radon. Though the VOA collection techniques involve extra handling of the sample, they can, however, produce results statistically similar to the syringe method, especially if a universal hose connector is used.

The data from the storage experiment indicated that loss of radon from VOA bottles could be a factor in some situations, but the loss resulting from radioactive decay has the greatest potential effect on storage time. Within 4 days, 50% of the radon originally present in a sample will be lost because of decay. For samples containing high levels of radon, permissible storage times could be substantial provided that the amount remaining at the time of analysis is above the practical quantification level. (For example, a sample containing approximately 7,670 pCi/L could be held up to 10 days, even with a 20% loss due to leakage, and still contain 1,000 pCi/L). However, the amount of radon in a sample is often not known, so the maximum storage time sufficient to obtain a valid measurement must be based on the MCL.

A hierarchical experiment was conducted using the direct syringe sampling technique and collection in VOA bottles with subsequent laboratory analysis. The total variability associated with the two methods was not significantly different ($\alpha \leq 0.10$, F test). The total variation of 4% to 6% as a result of sample handling and instrument variation was not high considering the volatile nature of radon. Most of the variability in the direct syringe technique (92.1% of total variance) was due to a combination of sample handling and instrumentation, whereas the variations due to sample handling and instrumentation for the VOA bottle technique were 55.5% and 44.5%, respectively.

An ANOVA showed that the mean count rates for the mineral oil-based and Opti-Fluor 0 cocktails were not significantly different ($\alpha \leq 0.10$); however, the mean count rate of the toluene-based cocktail was significantly less than both of these ($\alpha \leq 0.05$). In addition, the percent relative standard deviation (% RSD) associated with the toluene-based cocktail (3.22%) was greater than those of mineral oil-based (0.90%) and Opti-Fluor 0 (1.34%) cocktails. The choice of a scintillation cocktail may also be influenced by other factors such as cost, disposal, and mailing restrictions.

The EPA procedure for analyzing radon in water requires that the vial containing the sample be shaken to speed the extraction of radon from the water into the cocktail while other radionuclides remain in the aqueous phase. Both shaken and nonshaken radium-226 standards were analyzed in a series of experiments. The data indicated that transfer of the radon to the cocktail is continuous and does not require the extraction via shaking procedure, with the possible exception of samples containing very low radon activities. Use of efficiency factors, which account for time between extraction and counting, could lead to an underestimation of actual radon activities, especially for samples at or near the MCL.

Conclusions and Recommendations

When designing a treatment system to remove radon from a small community water supply, good data on water flowrates and influent radon activities at the site are essential. These data are major inputs into the design models for the aeration and GAC systems. As observed in this study, for small community supplies, variations in flowrate and influent activity may be substantial and will, if underestimated, lead to inadequate system design and effluent radon activities that exceed the design goal. Variations in water quality at each site may require that pilot-scale testing be performed to determine the appropriate design and pre- or post-treatment requirements.

For GAC systems, the steady state adsorption-decay constant, a critical component of the design model, varied over time and was site specific. The GAC systems tested in this study had average effluent radon activities of 12,000 to

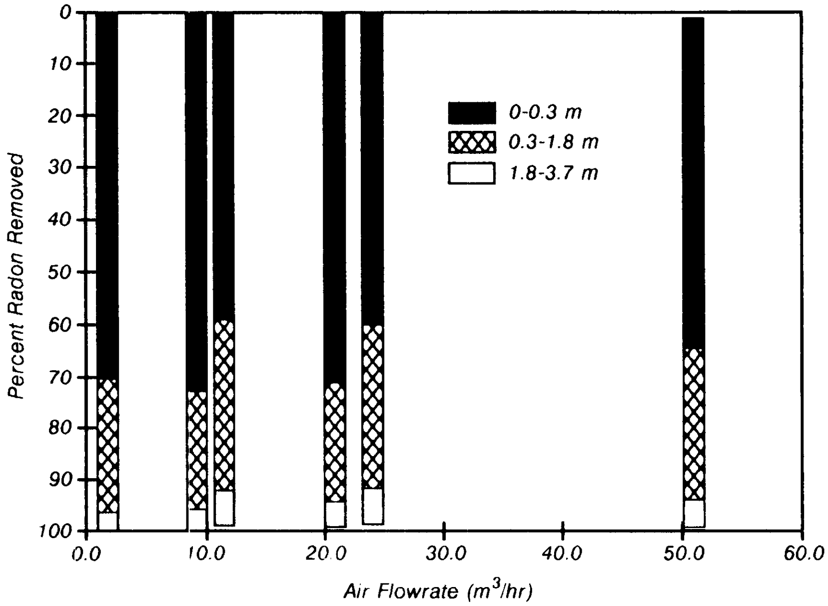


Figure 4. *Packed tower aeration – Mont Vernon, NH. Percent radon removal as a function of air flowrate within the tower for one packing type (mini rings).*

24,000 pCi/L. The GAC units accumulated iron, manganese and particulates (turbidity), and there were significant numbers of bacteria growing on the carbon. As a result, GAC systems may require periodic backwashing to prevent significant headloss development. In some cases, pretreatment is recommended to decrease the backwashing frequency. Gamma/beta emissions measured at the surface of the GAC units were substantially greater than background measurements possibly requiring shielding to lower them to acceptable levels. The data indicated that retention of uranium-238 and -235, radium-226, and lead-210, which appear to be related to water quality (e.g., pH and alkalinity), may cause the GAC to be classified as a low level radioactive waste.

The diffused bubble aeration system tested at A:W ratios ≥ 5:1 yielded overall radon removal efficiencies from 90.0% to >99.6%. The radon removal efficiencies for the packed tower aeration system ranged from 92.5% to 99.8%, in spite of variations in water flowrate, influent radon activity, and packing type. Extrapolations of performance data obtained at one site with either aeration system should not, however, be made to systems with other configurations, process equipment, or low influent activities, or to those required to meet a more stringent MCL. Both aeration systems had off-gas radon activities that were 10^4 to 10^5 times higher than those of the ambient air, and their effect on the environment would need to be considered. As with all aeration systems, precipitation of iron and manganese can occur and result in operational problems. Therefore, raw water quality should be monitored to determine whether pretreatment is required.

Several recommendations can be made concerning the liquid scintillation analytical technique for radon in water, During sample collection, the universal hose connector should be used to fill VOA bottles. Maximum storage times for samples collected in VOA bottles should be established based on the MCL, practical quantification lever, radioactive decay, and leakage. Opti-Fluor 0 yielded the best results of the scintillation cocktails tested with respect to count rates, variability, and cost and is, therefore, recommended. The extraction procedure should not be used to calculate the efficiency factor; however, samples should be shaken, especially those with low activities, to ensure rapid transfer of radon to the scintillation cocktail.

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Kim R. Fox *is the EPA Project Officer (see below).*
The complete report, entitled "Radon Removal Techniques for Small Community Public Water Supplies," (Order No. PB 90-257 809/AS; Cost: $31.00, subject to change) will be available only from:
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