Risk Reduction Engineering Laboratory Cincinnati OH 45268

Research and Development

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# **Project Summary**

# Radon Removal Using Point-of-Entry Water Treatment Techniques

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The report summarized here presents the results of a 1 yr evaluation of radon removal using point-of-entry (POE) granular activated carbon (GAC) adsorption with and without ion exchange pretreatment, diffused bubble aeration, and bubble plate aeration. The full report discusses each of the treatment alternatives with respect to their radon removal efficiency, potential problems (i.e., waste disposal, radiation exposure, equipment malfunctions, intermedia pollution), and economics.

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

In a recent status report, the U.S. Environmental Protection Agency (EPA) indicated that it is considering setting the Maximum Contaminant Level (MCL) for radon-222 (hereafter referenced as radon in the text) in the range 200 to 2,000 pCi/L. Several studies on the distribution of radon in groundwater supplies in the United States indicate that there will be a large number of private water supplies with activities in that range. Although an EPA regulation for radon would not apply to privately owned wells, states normally adopt Federal regulations and apply them to these water supplies. Private supplies exceeding the radon regulations will require POE treatment.

The purpose of this EPA Cooperative Agreement was to evaluate the performance of POE GAC and diffused bubble and bubble plate aeration systems while treating a groundwater supply containing radon. In this study, direct comparisons could be made among the individual POE systems because they were operated in a parallel flow configuration, each receiving the same influent water from an abandoned, smallcommunity groundwater supply. All systems treated 1.02 m<sup>3</sup>/day of water applied in a pattern designed to simulate daily demand for a POE system. Each of the treatment systems was evaluated with respect to three primary factors: radon removal efficiency, potential problems (i.e., waste disposal, radiation exposure, equipment malfunctions. intermedia pollution), and economics over a 1 yr period of operation.

Separate POE GAC systems were operated with and without ion exchange pretreatment and were monitored for changes in radon removal, radiation emissions, and general water quality parameters (e.g., pH, alkalinity, iron, manganese, turbidity, microbial numbers, and nonpurgeable dissolved organic carbon [NPDOC]). In addition, several special monitoring events were conducted on the GAC systems to assess the effect on performance of daily variations in water flowrate and raw water quality and backwashing. The ion exchange and GAC units were also cored after 1 yr of operation to determine if iron, manganese, microorganisms, or radionuclides (any or all of them) were accumulating within the bed. The POE diffused bubble and bubble plate aeration systems were monitored for radon

removal, general water quality parameters, and radon off-gas emissions.

## **Analytical Methods**

Standard Methods for the Examination of Water and Wastewater and EPA methods were used to determine radon; gamma/ beta emissions, the activity of total uranium. radium-226, and lead-210; microbial numbers; pH; temperature; turbidity; 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 instructions. Air monitoring using alpha track detectors was conducted outside the building housing the treatment units. The detectors were analyzed by Terradex Corp.\* (Glenwood, IL) after approximately 4 mo exposure.

### GAC POE Systems

The GAC units were designed by Lowry Engineering (Unity, ME), and both contained approximately 0.05 m³ of Barneby Cheney (Columbus, OH) Type 1002 activated carbon. Both GAC units were preceded by pleated paper sediment filters designed to remove particulates from the raw water. An ion exchange unit containing approximately 0.04 m³ of a strong cationic resin was installed between the sediment filter and the second GAC unit. The ion exchanger, which was regenerated every 2 wk, was designed to remove iron and manganese from the raw water before it entered the GAC.

#### Diffused Bubble POE System

The diffused bubble unit, designed by Lowry Engineering (Unity, ME), consisted of three small tanks in series, each containing a diffuser attached to a common header. The diffusers had variably spaced 0.64-mm-diameter holes. Water was pumped from the third compartment of the unit to a hydropneumatic tank. A timer connected to the blower was set to ensure that air flow continued 10 min after cessation of water flow. The exhaust, collected at the top of the unit, was vented outside the building.

#### Bubble Plate Aeration System

The bubble plate aeration unit was designed by North East Environmental Products (West Lebanon, NH). Raw water entered the unit directly from the well and was distributed by a spiral diffuser into a 7.6-cm-wide, 270-cm-long channel. The bottom of the channel was perforated by 4.8-mm-diameter holes spaced 1.9 cm apart. During

operation, air was forced up through these holes, and this caused the water column to rise to a height of 17 cm. At the end of the channel, water flowed over a weir into a holding tank; from there, it was pumped to a hydropneumatic pressure tank. A timer connected to the air blower was set to ensure that air flow continued for 1 min after cessation of water flow. Exhaust, collected from the top of the bubble plate chamber, was vented outside the building.

## Sampling Events

All systems were started in January 1989, and data collection was completed in January 1990. Approximately 1,022 L of water was distributed to each unit every day in one 30-min and six 18-min intervals at a flowrate of approximately 7.6 L/min. All units were monitored daily for the first 3 days. The GAC units were monitored every 3 to 4 days for the first month, weekly for the next 5 mo, and biweekly thereafter. The aeration units were monitored for the first 6 mo and biweekly thereafter.

During the loading rate studies, the flowrate to the GAC systems was between 7.6 and 20 L/min or the total daily throughput was changed from 1,022 L to 1,890 L, or both. The GAC units were also backwashed after 11 mo of operation using a water flowrate of approximately 7.6 L/min for 15 min.

## Data Analysis

All data were analyzed using Student's t tests at  $\alpha$  values of 0.01, 0.05, and 0.10. The most rigorous  $\alpha$  value of the three at which the stated trends occurred was reported for each data set tested.

#### **Results and Discussion**

The raw water characteristics at the test site are summarized in Table 1.

## Granular Activated Carbon

The sediment filters and ion exchange unit did not remove any detectable amount of radon from the water ( $\alpha = 0.05$ ). The activity in the effluent (Figure 1) was negligible for several days and then increased rapidly to an average of 881 ± 662 pCi/L for the GAC without pretreatment and 660 ± 204 pCi/L for the GAC with pretreatment; this activity continued for 4 mo. During the remaining 8 mo of operation, the effluent quality from both GAC units gradually decreased from a radon removal efficiency of 99.7% to between 79% and 85%. The radon mass loading applied to the GAC units was generally within the range expected and was least variable during the end of the study when radon removal decreased the most. Though the decrease in radon effluent quality started at approximately the same time that the NPDOC breakthrough occurred, the correlation between NPDOC and radon removal was not strong. Nor could the decreased efficiency be totally attributed to iron accumulation within the bed because both GAC units showed the same trend with respect to radon removal, although they received greatly different iron

Though the radon removal profile within the GAC units could not be determined directly, gamma emissions measurements were examined to determine where the zone of radon removal was occurring. Gamma radiation is emitted during the decay of bismuth-214 and polonium-214, which are short-lived progeny of radon. Hence, gamma emissions are highest where the greatest radon removal is occurring. Initially, the greatest gamma emissions occurred at the top of both GAC beds, but during the middle and end of the study, an equal or greater amount of emissions came from the mid-depth of the units (Figure 2).

Table 1. Raw Water Characteristics

Constituent	Concentration*		
Radon	35,620 ± 6,727 pCi/L		
рH	6.24 ± 0.19		
Temperature	11.8 ± 2.5°C		
Alkalinity	$34 \pm 19$ mg/L as CaCO,		
Calcium	$15.5 \pm 1.15  mg/L$		
Turbidity	1.04 ± 0.94 NTU		
Microbial Numbers	30,400 ± 29,500 CFU/100mL		
NPDOC'	$1.30 \pm 0.28  \text{mg/L}$		
Total Iron	$0.40 \pm 0.27  \text{mg/L}$		
Soluble Iron	$0.32 \pm 0.22  \text{mg/L}$		
Total Manganese	$0.36 \pm 0.22  \text{mg/L}$		
Soluble Manganese	$0.35 \pm 0.11  \text{mg/L}$		
Uranium	14.4 ± 6.5 mg/L		
Radium	3.5 ± 2.4 pCi/L		

<sup>&#</sup>x27;Mean ± Standard Deviation.

<sup>\*</sup>Mention of trade names or commercial products does not constitute endorsement or recommendation for use.

<sup>\*</sup>NPDOC = Nonpurgeable Dissolved Organic Carbon.

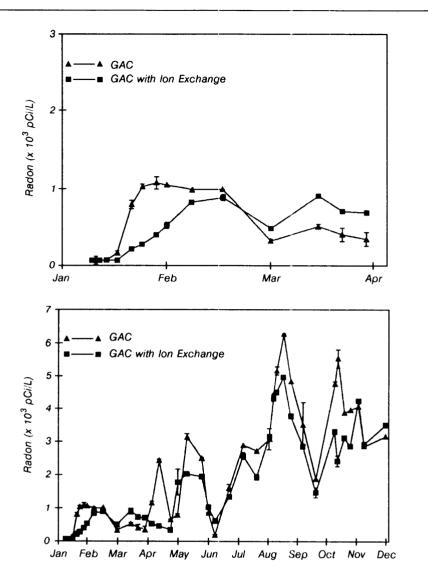


Figure 1. Effluent radon activities for the GAC systems for the first 4 months (a) and for the entire study (b). (Note the scale difference.)

These data indicated that the zone of radon removal was moving slowly down the beds. Because of the relatively low cost of purchasing 0.03 to 0.08 m³ of GAC, POE units usually contain a large amount of excess carbon that dampens potential overloads to the system and probably prolongs the life of the unit.

The lead-210 retention data (based on analysis of GAC core samples) and the gamma emissions data indicated that radon removal was occurring deeper within the GAC without pretreatment. This phenomenon appeared to be related to iron loading/accumulation in the bed. The effluent from the GAC without pretreatment contained no detectable iron (0.06 mg/L), whereas the

GAC with pretreatment received no detectable iron loading because the ion exchange unit preceded it. The iron data from the coring experiment of this unit showed that there was a significant accumulation ( $\alpha =$ 0.01) of iron precipitates (13.5 g/kg dry weight GAC) in the top of the GAC bed as compared with the GAC with pretreatment. During visual inspection of the GAC in the unit without pretreatment, a crust of orangebrown precipitate was observed in the top 15.2 cm of the bed. It is possible that the iron precipitation in the GAC impeded radon sorption by fouling the GAC surface or causing short circuiting (channeling) of water in the top of the bed. As a result, the radon removal front appeared to be moving

further down the bed in the GAC without pretreatment, so that the volume of excess carbon available for polishing was reduced.

Increases in loading rates to both GAC units resulted in increased radon activity in the effluent, especially for the GAC without pretreatment. The data indicated that, over time, the ability of GAC to dampen mass loading variations may be reduced. Backwashing did not change the radon removal observed in either GAC unit nor did it affect gamma profiles or any of the other water quality parameters monitored. There was a small, temporary decrease in effluent quality immediately after backwashing was completed, as is typically observed in granular media systems.

Gamma emissions measurements were taken 1.5 m away from the GAC tanks (Figure 3) and were within 80% to 100% of the values predicted using the Carbdose 2.0 model developed by B. Keene and S. Rydell (USEPA Region I; Boston, MA; 1989). At the end of the study, the GAC with pretreatment was placed in the center of a 61-cm-diameter plastic tank filled with water. The data indicated that the water jacket attenuated the gamma radiation by 14% to 17% at a distance 30.5 cm away from the GAC unit.

Uranium was removed from the water as it passed through the GAC without pretreatment (80% ± 32% removal), the ion exchanger (73% ± 23%), and GAC with pretreatment (35%  $\pm$  39%). At a raw water pH of 6.03 ± 0.34, the predominant uranium species is the neutral uranyl carbonate complex, UO,CO,, which should be adsorbed by the GAC. There was good agreement between the coring data and the uranium removal based on measurements of the treated water. According to the draft "EPA Guidelines for Disposal of Drinking Water Treatment Plant Residues Containing Naturally Occurring Radionuclides." the GAC would fall into the range 30 to 300 pCi/ g and, therefore, could be placed in a stabilized landfill. The ion exchange resin would not require specialized treatment (approximately 14 pCi/g) with respect to uranium.

A small amount of radium removal was detected through the GAC without pretreatment, whereas the GAC with pretreatment did not receive any detectable input of radium (<0.10 pCi/L) because it was preceded by the cationic exchange resin. The coring data also indicated that little radium was retained by the GAC, which was expected because radium is extremely hydrophillic. Based on the draft EPA guidelines, the GAC from both units could be disposed of without specialized treatment with respect to radium because it contained <3 pCi/g. The ion exchange resin, even after regeneration, would require disposal

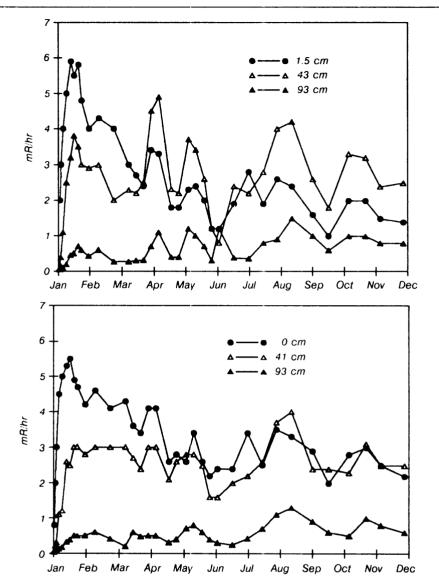


Figure 2. Gamma emissions measurements taken on the unit's surface at various depths for the GAC without pretreatment (a) and the GAC with pretreatment (b).

in a stabilized landfill because it retained between 10 and 22 pCi/g of radium.

The GAC retained 65 to 445 pCi/g of lead-210; this falls in the draft EPA guidelines range of 30 to 2,000 pCi/g and requires disposal in a stabilized landfill. Lead-210 was not retained by the ion exchange unit; this was not surprising because the unit did not remove radon from the raw water.

In addition, the ion exchange resin was contaminated with many other radionuclides probably retained from the raw water or removed by adsorption or ion exchange reactions occurring with other solid phases (e.g., Fe(OH)<sub>3</sub>, MgCO<sub>3</sub>) or organic matter. The water exiting the ion exchange unit during brine regeneration contained substantial (10<sup>2</sup> to 10<sup>3</sup> pCi/L) amounts of uranium and radium. Disposal of this slurry

should be carefully evaluated to determine the appropriateness of any disposal alternative.

## Aeration Systems

The diffused bubble unit consistently produced effluents with radon activities <200 pCi/L (Figure 4), even when the air flowrate was restricted because the diffusers became encrusted with iron precipitates. Removal efficiencies of >99% were expected because the influent radon activity was not exceptionally high, the air:water (A:W) ratio was large (approximately 119:1), and the bubble size was relatively small.

The bubble plate unit (Figure 4) also produced a very high quality effluent (>99% radon removal), except on a few occasions when the unit experienced mechanical

problems (e.g., solenoid valve and pump failure) and when the air intake filter for the blower was clogged (August-December). The A:W ratio for the bubble plate unit was also very high (approximately 156:1).

The pH of the effluents from both aeration units (diffused bubble =  $7.3 \pm 0.4$ ; bubble plate =  $7.2 \pm 0.4$ ) were not significantly different from each other ( $\alpha$ =0.10), but were significantly ( $\alpha$ =0.01) higher (by approximately 1.0 pH unit) than the influent. The increased pH probably resulted when carbon dioxide was removed from the water.

The effluent total iron concentrations for the bubble plate  $(0.43\pm0.24~\text{mg/L})$  and the diffused bubble  $(0.53\pm0.30~\text{mg/L})$  units were not significantly  $(\alpha=0.10)$  different from the influent concentration. The concentration of soluble iron, however, decreased significantly  $(\alpha=0.01)$  as almost all of the soluble iron was oxidized and precipitated by the aeration process. There was a concurrent increase in turbidity exiting the aeration units that correlated strongly to the increase in particulate iron.

Air monitoring was conducted to determine if the exhaust from the aeration units produced a detectable effect on the ambient air radon activities surrounding the building housing the units. Alpha track detectors located 1.5 to 24 m away from the pumphouse all registered <0.3 pCi/L (the detection limit). The detectors located 1.35 m directly under the vents for the aeration units registered 1.4 to 2.1 pCi/L. Considering the radon exiting the vents was in the range of 200 to 300 pCi/L, venting the exhaust from these POE units above a home's roofline should be sufficient to obtain adequate dilution of the radon plume to ambient air levels.

#### **Economics**

Conventional engineering economic practices were used to obtain a detailed economic evaluation of the technologies examined in this project. The economic evaluation assumed all water system components (e.g., well, pressure tank, piping) would already be in place and therefore, only the costs related to installation and operation of radon removal systems are presented. For comparative purposes, annual costs and production costs were developed for each treatment system. The production costs were expressed in conventional units of \$/1,000 gal to be consistent with those reported in other studies. The annual cost was computed as the sum of the amortized capital cost and the operation and maintainence cost. The amortized capital cost is the total capital cost amortized over a 5 yr time period at a 9% interest rate. A 5 yr time period reflects a typical home equity loan (i.e., typical means of

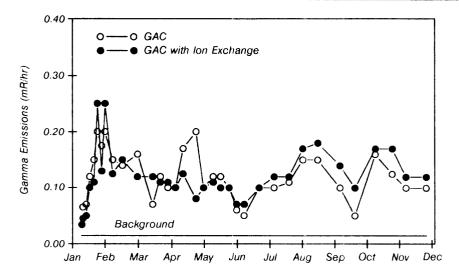


Figure 3. Gamma emissions measurements for the GAC systems taken 1.5 m away from the units' surfaces.

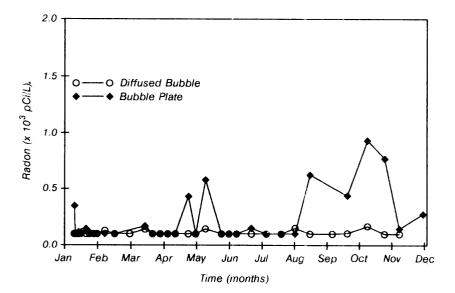


Figure 4. Effluent radon activities for the diffused bubble and bubble plate aeration units.

financing POE costs) payback period. All cost figures were updated to first quarter 1990 dollars using the ENR Construction Cost Index (CCI). The 1967 base year CCI index for first quarter 1990 is 435.

The cost estimates presented in the report are intended to give a general indication of the economics of the POE radon removal technologies studied. Specific treatment requirements will vary from site to site and may not be accurately reflected by the systems or assumptions used in the economic analysis.

Based on a design flow of 1.02 m³/day, the production costs for GAC without and with ion exchange pretreatment were \$9.31/

1,000 gal and \$12.25/1,000 gal, respectively. If the GAC required disposal as a low level radioactive waste, the costs would increase to \$9.88/1,000 gal and \$13.40/1,000 gal, respectively. These costs would be lower if the GAC and ion exchange resin could be disposed of in a stabilized landfill.

Total production cost estimates for the diffused bubble and bubble plate aeration systems were \$22.58/1,000 gal and \$26.74/1,000 gal, respectively. The major discrepancy between the costs of the two aeration systems resulted from the differential in the retail price of the equipment. (The costs of the aeration units did not include any special off-gas treatment or auxiliary blowers

but did include piping the radon vent above the roofline).

The production costs for the POE radon removal systems are high when compared with public water supplies because (a) there is no economy of scale and (b) it is assumed individual homeowners will be purchasing single POE units with short-term home equity loans unlike utilities that could purchase large quantities of POE units for a service area with a long-term loan. Therefore, there will not be a quantity discount on the POE equipment for the individual well water supplies nor a long amortization period.

## Conclusions and Recommendations

Of the three systems evaluated, the GAC requires the least owner maintenance, is the easiest to operate, and is the least expensive with respect to capital and operation and maintenance costs, even if low level radioactive waste disposal is required. Neither GAC system, however, consistently produced effluent radon activities in the range of the proposed MCL (200 to 2,000 pCi/L). The data indicate that the only condition where POE GAC systems could produce effluent in this range would be if the influent activity were low, requiring <80% removal. Other researchers have shown that when the influent radon activity is low (i.e., <5,000 pCi/L), the gamma emissions from the GAC units would yield acceptable levels of exposure dose.

It appears that retention of iron precipitates could shorten the life of the GAC with respect to radon removal and make it more susceptible to changes in loading. In applications where iron and manganese concentrations are high, ion exchange pretreatment may be essential. This presents problems, however, because the resin becomes contaminated with long lived radionuclides and the heavily contaminated regenerant brine and backwash water may require special disposal. In cases where frequent backwashing of the GAC and use of a sediment filter can limit accumulation of particulates and metal precipitates in the GAC bed, ion exchange pretreatment should be avoided.

One of the ancillary problems associated with GAC treatment is the retention of longer lived radionuclides in quantities that dictate the carbon must be disposed of in a special manner. Even if the quantities of the longer lived radionuclides, such as radium, are low in the raw water, lead-210 will always accumulate in a GAC unit retaining radon. A more thorough evaluation must be made to determine whether the benefits of GAC are outweighed by the problems associated

with specialized handling and disposal, when the raw water radon activity is low.

Both the bubble plate and diffused bubble POE units were very efficient (>99%) at removing radon from the water, even when it was present at relatively high levels. Therefore, these units should be able to meet an MCL of 200 to 2,000 pCi/L over a wide range of loadings, primarily because of the high A:W ratios used.

Of the several problems with aeration, iron oxidation is the most prominent. Even at low iron concentrations, precipitates can form and accumulate in the units, or be released in pulses to the residence when the unit is started to meet demand, or both. Iron treatment will probably be required to avoid these problems. In the diffused bubble unit, pretreatment may be necessary to prevent the diffusers from clogging. If ion

exchange pretreatment is used, however, the problems associated with radionuclide retention would have to be addressed. Post treatment of iron precipitates using sand filtration has been used, but a thorough investigation of these units in POE applications should be performed before their widespread installation.

In addition to having higher capital and operation and maintenance costs than POE GAC systems, the aeration units are more prone to operational problems because they have more mechanical parts. As a result, frequent maintenance will be essential to the proper operation of the aeration units. Proper venting of the off-gas from the units will require discharge above the roofline of the home.

Elevated levels of bacteria (up to 200,000 CFU/100 mL) were periodically obtained in

the effluent from all of the POE units tested. Depending on EPA and state regulations, it may be necessary to disinfect the treated water before consumption. Finally, because radon cannot be readily detected by the senses, there is a potential for it to be reintroduced into the water supply if a POE unit fails without the residents being aware of the problem. Therefore, frequent monitoring of the effluent from any of the POE units should be stressed to the homeowner.

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Kim R. Fox is the EPA Project Officer (see below).

The complete report, entitled "Radon Removal Using Point-of-Entry Water Treatment Techniques," (Order No. PB91-102 020/AS; Cost: \$23.00 subject to change) will be available only from:

National Technical Information Service

5285 Port Royal Road Springfield, VA 22161 Telephone: 703-487-4650

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