NOTE: Fig. 2 & 3 y axes should be "nCi/L" Fig. 4 y axis should be "ln Radon, pCi/L"

Modeling Point-of-Entry Radon Removal by GAC

Jerry D. Lowry and Sylvia B. Lowry

Results of a study which showed that an adsorption–decay steady state is established within a granular activated carbon (GAC) bed, allowing it to be maintenance-free for long periods of time, led to field research to develop a design model for point-of-entry application and the installation and monitoring of GAC technology in more than 100 homes across the United States. The research showed that with an effective GAC better than 99 percent removal of radon can be achieved.

The health implications of airborne radon-222 (^{222}Rn) in households are well documented,[1-8] as is the significance of an elevated ^{222}Rn level in water and how it contributes to the airborne ^{222}Rn concentration.[1,2,9-19]

The feasibility of removing ^{222}Rn from household and small system water supplies with granular activated carbon (GAC) or aeration devices has been reported by various researchers,[1,20-23] and a detailed report regarding radon removal from public water supplies has recently been prepared for the US Environmental Protection Agency (USEPA).[24]

In response to increasing concern about airborne and waterborne ^{222}Rn, research was initiated at the University of Maine in 1980 to identify technologies to remove ^{222}Rn from groundwater.[17,22] Aeration and GAC were identified as potentially cost-effective treatment processes for point-of-entry applications. Laboratory testing of full-scale point-of-entry GAC and diffused bubble aeration units showed that both methods were effective, but GAC appeared more promising for household applications. That study and a previous one documented that an adsorption-decay steady state is established within a GAC bed, allowing it to be virtually maintenance-free for an indefinite but long period of time. These studies led to laboratory research to model the GAC process for municipal application, field research to develop a design model for point-of-entry application, and installation and monitoring of the GAC technology in more than 100 households throughout the United States. This article reports on the findings of the second and third aspects of the preceding research.

Development of a GAC model

Previous research indicated that the adsorption-decay steady-state performance could be modeled by first-order kinetics, allowing the use of the following equation to describe and predict removal:

$$C_t = C_o e^{-K_{ss}t}$$

in which C_t = the ^{222}Rn concentration at time t in picocuries per litre, C_o = the

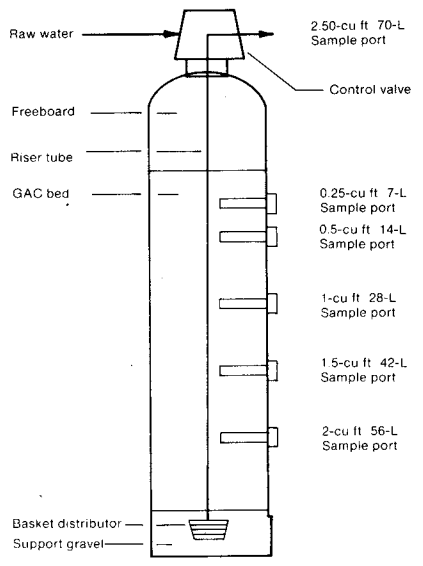


Figure 1. Experimental GAC vessel for K_{ss} determination

TABLE 1
Summary of adsorption-decay design constants

Carbon	Manufacturer	Type	K_{ss}—h^{-1}
A	American Norit	Peat (8 × 20)	1.35
B	ICI Americas*	HD4000 (12 × 40)	2.09
C	Calgon	F-400 (12 × 40)	1.53
D	Barneby Cheney	299 or 1002	3.02

*Now manufactured by American Norit

TABLE 2
Typical levels of uncertainty for this study

^{222}Rn Concentration *pCi/L*	Uncertainty *percent*
800,000	0.5
300,000	0.5
40,000	1.0
2,000	4.5
1,000	6.0
500	15
100	25
60	45

initial ^{222}Rn concentration in picocuries per litre, K_{ss} = the steady-state adsorption-decay constant per hour, and t = the length of empty bed detention time (EBDT) in hours.

This is logical because at steady state the adsorptive removal equals the decay of adsorbed ^{222}Rn at all points in the bed, and decay kinetics are first order. The actual achievement of the adsorption-decay steady state is quite complex. It involves non-steady-state adsorption kinetics, which dominate the early period, followed by an increasingly significant decay phenomenon set up by ^{222}Rn and its short-lived progeny. The result of these processes is the establishment of the steady-state distribution of ^{222}Rn and its short-lived progeny on the GAC. Although the establishment of the steady state is a complex process, the performance after steady state is reached can be accurately described by the simple steady-state model.

The use of the average EBDT is appropriate because the GAC bed at steady state acts as a decay-storage device. This means that for all practical purposes, the normal intermittent diurnal flow experienced in a typical household is not important in terms of its effect on the steady-state performance. An analogy can be made to a chromatographic column in which the ^{222}Rn travels along the bed at a relatively slow rate compared with the water and decays down to the effluent value. The bed is simply a concentrating device that stores ^{222}Rn and is equivalent to a plug flow storage tank having a much greater liquid detention time. For example, a GAC bed giving a 99 percent reduction is equivalent to an ideal plug-flow decay-storage tank having a detention time of 25.3 days. Thus, t should be calculated for the water volume used over a period of two to three weeks because this is the period to which the GAC bed is responding. In a typical household, the three-week flow average does not vary significantly, except perhaps seasonally in a gradual manner.

To accurately document this model and to test the relative effectiveness of several different GAC products, a field study was designed to measure the steady state adsorption-decay constant, K_{ss}. A Maine household with extremely high levels of ^{222}Rn in its groundwater supply was selected to demonstrate the general applicability of the model across a wide range of ^{222}Rn levels. The performance of three GAC products (Table 1) was examined using a modified commercial treatment unit (Figure 1). Slotted (0.012-in. [0.3-mm]) laterals were installed at depths of 0.25, 0.50, 1, 1.5, and 2 cu ft (7, 14, 28, 42, and 57 L) bed volumes to obtain depth samples. The total bed volume was 2.5 cu ft (71 L).

Water samples for ^{222}Rn analysis were taken directly from sampling valves with a 10-mL syringe, which was subsequently discharged directly into a previously prepared liquid scintillation vial containing 5 mL of fluor. The vial was immediately capped and mailed to the laboratory for counting. The basic counting procedure used was one described by Pritchard and Gesell,[25] except that a mineral oil rather than a toluene-based fluor was utilized because of postal regulations. Precision of the ^{222}Rn analysis is a function of the level of ^{222}Rn present, the counting time, and the time elapsed between sampling and counting. Typical levels of uncertainty (2-sigma) for this study are given in Table 2.

The water use at the household was monitored by a standard ⅝-in. (17-mm) meter and totalizer readings were taken daily. Water temperatures ranged between 43 and 50°F (6 and 10°C) throughout the study.

The various GAC products were tested sequentially, each by the same method. The virgin carbon was placed in the pressure vessel over a gravel support and commissioned after a backwashing period of 15 to 30 min to remove fines. The GAC bed remained in service to allow a steady state to be achieved (approximately three weeks) and was monitored for an additional three- to four-week period. Fourteen sets of samples (all ports) were collected to determine K_{ss}.

Typical examples of the results of the field testing are illustrated by Figures 2-4 for GAC B. Figures 2 and 3 show the results for the depth of ^{222}Rn removal and the establishment of the adsorption-decay steady state. The exact reason for the elevated point for each depth on day 42 was not known, but was suspected to be the result of desorption because of extreme raw water ^{222}Rn variation that was not documented by sampling the previous week. This particular well is subject to such variations, and previous monitoring has documented that the ^{222}Rn variation over a period as short as several days can be from 150,000 to more than 2,000,000 pCi/L. For bed volumes deeper than 1.5 cu ft (42 L), these variations are not significant in relation to the raw water concentrations because there is enough GAC to provide adequate dampening. They are apparent in this case because of the extremely high average raw water ^{222}Rn.

A semilogarithmic plot of bulk solution ^{222}Rn (ln) versus EBDT yields a linear relationship with a slope equal to K_{ss}. This relationship is illustrated in Figure 4 for one of the carbons tested.

The analysis of variance (ANOVA) for the data is summarized in Table 3. Although the deviations from regression were statistically significant, they were small compared with the variation explained by regression. This fact is reflected by the relatively narrow confidence limits (95 percent) around the least-squares regression line in Figure 4.

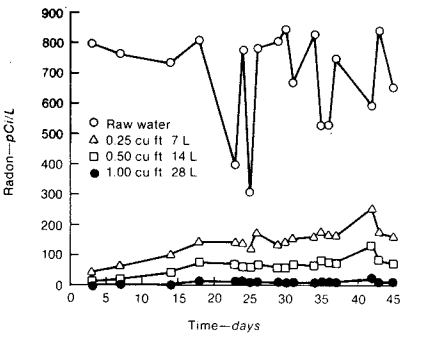


Figure 2. Performance for the top portion of the GAC bed for GAC B

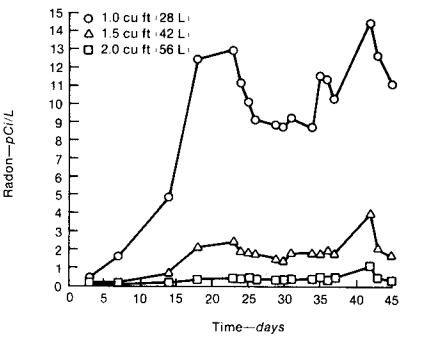


Figure 3. Performance for the bottom portion of the GAC bed for GAC B

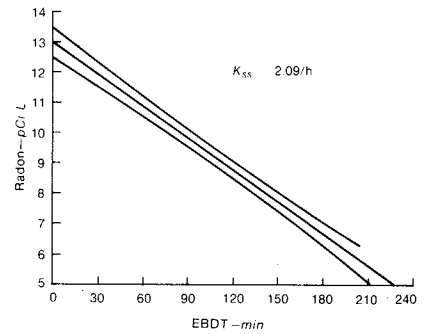


Figure 4. First-order steady-state adsorption-decay relation for GAC B, with 95 percent confidence limits indicated

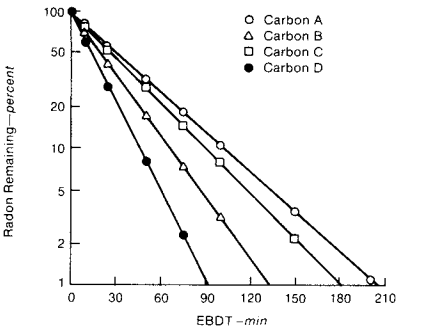


Figure 5. Contrast of the steady-state adsorption-decay relation for test carbons A, B, and C with GAC D

TABLE 3
Summary of analysis of variance for GAC B

Source of Variation	*df*	*SS*	*MS*	*F*
Among groups (EBDT)	5	534.7	106.9	829*
Linear regression	1	530.6	530.6	515*
Deviations from regression	4	4.1	1.0	8.0*
Within groups (ports)	78	10.1	0.13	

*Significant at 0.001 level

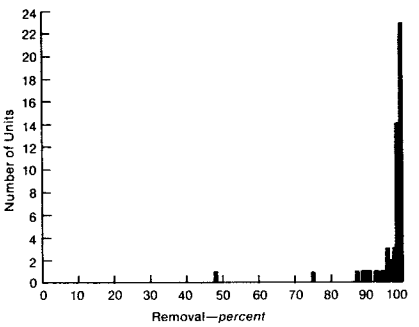


Figure 6. Histogram of the steady-state performance of 66 GAC treatment units

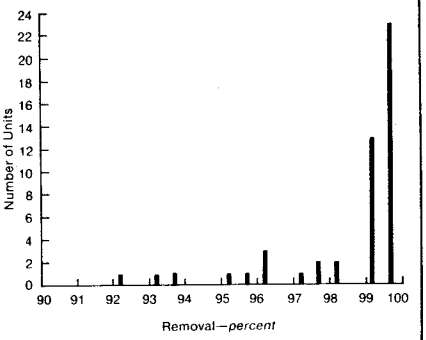


Figure 7. Histogram of the steady-state performance of properly operating GAC units containing GAC D

The performance for carbons A, B, and C are contrasted with that of carbon D, which has been determined by other researchers[23,26] to have the highest K_{ss} value tested to date. A summary of the K_{ss} values for these carbons is given in Table 1. The ranking of these carbons bears little relation to how they performed previously by isotherm testing and shows that isotherms are not indicators of how a carbon will perform at steady state.[17] This group of four carbons contains the best and worst carbons, with respect to their ability to remove ^{222}Rn. It is clear that the type of GAC selected has significant bearing on the performance achieved. For example, at 99 percent removal the required carbon volume for carbon D is 50 percent of that needed with carbon A. For equal bed volumes, carbon D achieves a 99 percent reduction, compared with an 88 percent reduction with carbon A. The bulk densities for carbon A and D are approximately 18 and 32 lb/cu ft, respectively, and on a mass rather than a volume basis, all GAC types tested are much closer in performance. Because the number and size of vessels required are determined by the volumetric performance, this has little practical significance. Of more importance is the probable positive influence of decreasing particle size. No studies have documented, however, the magnitude of this factor for the ^{222}Rn steady state.

Because of the small flow treated and the relatively narrow range of bed sizes required to cover the entire range of ^{222}Rn encountered in point-of-entry applications, the long EBDT is easily satisfied by commercially available pressure vessels. A range of bed volumes from 1 to 3 cu ft (28 to 85 L) will remove more than 98 percent of the ^{222}Rn from any household groundwater supply. In contrast, the relatively long EBDT is of significant importance when increasing design flow in municipal applications. Compared with an EBDT of approximately 15 min for organics removal, the residence time for ^{222}Rn removal is quite long. Although the GAC will last indefinitely in the ^{222}Rn application, the initial high capital cost for the GAC makes aeration an attractive alternative for larger water systems.

Field experience with GAC treatment

Since 1981, GAC units have been installed in a number of households to remove ^{222}Rn from water and thereby lower the airborne ^{222}Rn levels in homes. The current number of units that exist for this purpose is unknown but is estimated to be in excess of 500. Approximately 100 units have been installed and monitored as part of a data base for future research on aspects other than simple removal, such as the resulting gamma exposure rate from ^{214}Pb and ^{214}Bi and the long-term buildup of ^{210}Pb. In each of these installations, the GAC type, the GAC quantity, and the installation date are known. In some of the installations the water use is known, and the radium and uranium content of the raw water has been documented. This data base is unique in that it covers GAC units that have been installed over a wide area—12 states and Canada—and includes the longest operating GAC units for ^{222}Rn. In addition, these units are installed on water supplies with less than 1,000 pCi ^{222}Rn/L to more than 1,000,000 pCi ^{222}Rn/L.

Although four GAC types have been used in these units, more than 85 percent contain carbon D and 10 percent contain carbon C. A summary of the steady-state performance for all routinely monitored installations is presented in Figure 5. With the exception of three units (in which GAC channeling may have resulted from horizontal shipping of the unit and shifting of the accompanying support gravel in transit), the performance level in field installations is very high. Eighty percent of all units are in the 1.7-cu ft (48-L) category, with remaining units ranging from 1 to 3 cu ft (28 to 85 L). The average removal of ^{222}Rn for all units is 96.2 percent.

Elimination of three known malfunctioning (channeling) units and the units containing carbon C yields the histogram summarized in Figure 6. For these units, the average removal of ^{222}Rn is 98.9 percent. It is uncommon to monitor a unit and find less than 99 percent removal. Although the real value of these units lies in future studies involving ^{222}Rn progeny buildup, these data demonstrate the high degree of removal that can be obtained with properly designed and installed systems.

GAC versus aeration

It has been documented that aeration is a feasible method for ^{222}Rn removal in point-of-entry applications.[22,27] But a number of factors have prevented it from becoming as popular a method as GAC treatment.

- Aeration is performed at atmospheric pressure and, therefore, requires repressurization of the water supply.
- The initial cost of aeration systems designed for ^{222}Rn removal is relatively high, partly because of the repressurization requirement. The installed cost ranges from $2,500 to more than $4,000, compared with approximately $650 to $1,200 for GAC.
- Unlike GAC, several aeration methods have limited removal capabilities. A novel but costly spray aeration system was developed by the Maine Department of Human Services, Division of Health Engineering. Six units are operating in the field. These units achieve 90 to 95 percent removal, but may be limited to wells containing only 10,000 to 20,000 pCi/L if, for example, the future USEPA maximum contaminant level (MCL) for

^{222}Rn is set at 1,000 pCi/L. Although the MCL would apply only to public water supplies, the real ^{222}Rn issue is in private household supplies—in many cases, lending institutions are already requiring removal. These institutions tend to use the MCLs as guidance for private supplies as well.

Packed-tower aeration systems are confined by the ceiling height in the cellar or living area and are therefore limited to about 90 percent removal. A multistaged diffused bubble aeration system developed for organics removal[28] was tested on a supply that contains 250,000 pCi/L and removed ^{222}Rn to below detection, for virtually 100 percent removal. However, even a less expensive version, designed specifically for ^{222}Rn removal, would cost about two to three times as much as a GAC unit.

- Aeration methods have significant operation and maintenance requirements, which increase the cost differential over the long term.

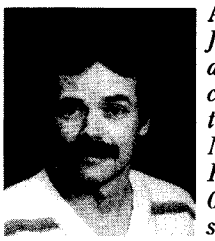
Aeration methods have a single advantage to be considered for point-of-entry applications, i.e., they avoid the elevated gamma exposure rate and the possible long-term buildup of ^{210}Pb associated with GAC beds. These topics are currently the subjects of ongoing research and are beyond the scope of this article. However, economical shielding and proper location of the treatment unit can eliminate any potential gamma exposure in excess of normal background levels. The buildup of ^{210}Pb in these applications is not documented, but it could be a concern. Future documentation and research on this subject will determine the extent of and solutions to these problems. If, however, a more economical aeration system is developed that is capable of 99 percent removal, it may generally be applied along with GAC systems for ^{222}Rn removal in point-of-entry applications.

Conclusions

- A first-order model accurately describes the adsorption-decay steady-state removal of ^{222}Rn by GAC.
- A single design constant, K_{ss}, can be used to rank a given GAC type for ^{222}Rn removal. The ranking of a carbon for steady-state performance does not appear to be related to its ranking according to an adsorption isotherm.
- There is a significant range of the design constant K_{ss} for the carbons tested to date, making the selection of the correct GAC important. This is especially true for small public water supply applications, in which vessel size is more sensitive to economics.
- More than 99 percent reduction of ^{222}Rn is possible with an effective GAC.
- The progeny of ^{222}Rn make it important to consider the location of and protective shielding for a GAC bed, to avoid increased gamma exposure over normal background levels.
- The potential for the long-term accumulation of ^{210}Pb in GAC beds should be investigated.

References

1. CASTREN, O. The Contribution of Bored Wells to Respiratory Radon Daughter Exposure in Finland. Tech. Rept., Inst. of Radiation Protection, Helsinki, Finland (1977).
2. HESS, C.T. ET AL. Radon in Potable Water Supplies in Maine: The Geology, Hydrology, Physics, and Health Effects. Completion Rept., Land and Water Resources Ctr., Univ. of Maine and Ofce. of Water Res. and Technol., US Dept. of the Interior. Washington, D.C. (1979).
3. EVANS, R.D. ET AL. Estimate of Risk From Environmental Exposure to Radon-222 and its Decay Products. *Nature,* 290:98 (1981).
4. HARLEY, N.H. Editorial—Radon and Lung Cancer in Mines and Homes. *New England Jour. Med.,* 310:23:1525 (June 7, 1984).
5. NAS Comm. on Biological Effects of Ionizing Radiation. The Effects on Populations of Exposure to Low Levels of Ionizing Radiation. Natl. Acad. Press, Washington, D.C. (1980).
6. USEPA. A Citizen's Guide to Radon: What It Is and What To Do About It (Aug. 1986).
7. USEPA. Radon Reduction Methods: A Homeowner's Guide, OPA-86-005 (Aug. 1986).
8. COTHERN, R.C. Estimating the Health Risks of Radon in Drinking Water. *Jour. AWWA,* 79:4:153 (Apr. 1987).
9. CASTREN, O. ET AL. High Natural Radioactivity of Bored Wells as a Radiation Hygienic Problem in Finland. Presented at the 1977 Intl. Radiation Protection Assn. Fourth Intl. Congress. Paris, France.
10. DUNCAN, D.L.; GESELL, T.F.; & JOHNSON, R.H. Radon-222 in Potable Water. Proc. 10th Midyear Topical Sym.: Natural Radioactivity in Man's Environment. Health Physics Soc., Saratoga Springs, N.Y. (Oct. 1976).
11. HESS, C.T. ET AL. Investigation of Rn-222, Ra-226, and U in Air and Groundwater of Maine. Completion Rept. B-017-ME, Land and Water Resources Ctr., Univ. of Maine and Ofce. of Water Res. and Technol., US Dept. of the Interior. Washington, D.C. (1981).
12. PARTRIDGE, J.E.; HORTON, T.R.; & SENSINTAFFER, E.L. A Study of Radon-222 Released During Typical Household Activities. ORP/EERF-79-1, USEPA, Ofce. of Radiation Prog., Eastern Envir. Radiation Facility, Montgomery, Ala. (Mar. 1979).
13. WEIFFENBACH, C.V. Radon in Air and Water: Health Risks and Control Measures. Technol. Transfer Rept., Land and Water Resources Ctr., Univ. of Maine (June 1986).
14. HORTON, T.R. Methods and Results of EPA's Study of Radon in Drinking Water. EPA 520/5-83-027, USEPA, Ofce. of Radiation Protection Programs—Eastern Environmental Radiation Facility. Montgomery, Ala. (Dec. 1983).
15. ALDRICH, L.K.; SASSER, M.K.; & CONNERS, D.A. Evaluations of Radon Concentrations in North Carolina Ground Water Supplies. Dept. of Human Resources Div. of Facility Services—Radiation Protection Branch. Raleigh, N.C. (Jan. 1975).
16. LOWRY, J.D. & MOREAU, E. Removal of Extreme Radon and Uranium From a Water Supply. Proc. of the 1986 National Conf. on Envir. Engrg., Envir. Engrg. Div., ASCE. Cincinnati, Ohio (July 1986).
17. LOWRY, J.D. & BRANDOW, J.E. Removal of Radon From Water Supplies. *Jour. Envir. Engrg. Div.—ASCE,* 111:4 (Aug. 1985).
18. LOWRY, J.D. Radon At Home. *Civil Engrg.,* 57:2 (Feb. 1987).
19. PRITCHARD, H.M. The Transfer of Radon From Domestic Water to Indoor Air. *Jour. AWWA,* 79:4:159 (Apr. 1987).
20. HOATHER, R.C. & RACKHAM, R.F. Some Observations on Radon in Waters and Its Removal by Aeration. Proc. Inst. Civil Engrs., London, England (Dec. 7, 1962).
21. SMITH, B.M. ET AL. Natural Radioactivity in Ground Water Supplies in Maine and New Hampshire. *Jour. AWWA,* 53:1:75 (Jan. 1961).
22. LOWRY, J.D. ET AL. Point-of Entry Removal of Radon From Drinking Water. *Jour. AWWA,* 79:4:162 (Apr. 1987).
23. LOWRY, J.D. Extreme Levels of ^{222}Rn and U in a Private Water Supply. Proc. Conf. on Radon, Radium, and Other Radioactivity in Ground Water: Hydrologic Impact and Application to Indoor Airborne Contamination. NWWA., Somerset, N.J., April 7-9, 1987.
24. Malcolm Pirnie Inc. Technologies and Costs for the Removal of Radon From Potable Water Supplies. Draft of report to USEPA under contract 68-01-6989 (Jan. 6, 1987).
25. PRITCHARD, H.M. & GESELL, T.F. Rapid Measurements of Radon-222 Concentrations in Water With a Commercial Liquid Scintillation Counter. *Health Physics,* 33:577 (1977).
26. PINNETTE, J. Master's thesis. Univ. of Maine, Orono (1985).
27. HOXIE, D. Personal communication (Mar. 1987).
28. LOWRY, J.D. & LOWRY, S.B. Restoration of Gasoline-Contaminated Household Water Supplies to Drinking Water Quality. Proc. Eastern Regional Ground Water Conf., NWWA, Portland, Maine (July 1985).



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