GRANULATED ACTIVATED CARBON WATER TREATMENT and POTENTIAL RADIATION HAZARDS

By
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INTRODUCTION

Almost thirty years ago Smith et al. [1] reported high levels of radon in the ground waters of northern New England. Twenty years later, concerns about the health effects of radon and the presence of high radon concentrations in some ground waters of Maine led to studies by a number of researchers, many associated with the University of Maine at Orono [2-5]. Early in these investigations it became clear that 1) radon is readily released from water through normal household water use and 2) the principal health risk from radon is from inhalation rather than ingestion. Consequently, indoor use of untreated water with a high radon concentration leads to an unnecessary risk of lung cancer. Reduction of this risk is the goal of the proposed regulation of radon in public water supplies by the Environmental Protection Agency. It is becoming increasingly common in New England real estate transactions to transfer to private or domestic water supplies the regulations protecting public health as stated in the Safe Drinking Water Act.

Although the results of investigations reported here are specific to radon (Rn-222) removal from domestic water supplies, the principles and problems apply to all systems where treatment is by granular activated carbon (GAC), regardless of the contaminant being removed. The subjects of this paper are the secondary problems of gamma radiation and disposal of radionuclides resulting from the accumulation of radioactive radon progeny on GAC filters.

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GAC Adsorption

Early in the investigation of radon treatment technology it became apparent that two basic methods, 1) aeration by various techniques and 2) granular activated carbon filtration were both better than 95% effective for reducing radon in water [6].

The technical aspects of the adsorption of radon on GAC have been extensively described by Lowry et al., and the practical field application to domestic and small public water supplies reported [8,9,10].

An important point for the present discussion is that a radon adsorption/radioactive decay steady state relationship is attained in a GAC bed after about thirty days. As a result of this steady state condition and the very small mass of matter retained by the GAC, a GAC bed is theoretically capable of constant and essentially perpetual radon removal. Uranium and, to a lesser extent, radium are also removed from water by GAC filters; but this will not be discussed here.

The appeal of GAC over aeration for very small systems has been its relatively low initial cost coupled with low operation and maintenance costs. This apparent advantage of the GAC treatment system is a result of its simple design, the lack of moving parts and no need to repressurize for distribution. Depending on the initial radon content and the percent radon removal required to meet a given finished water quality, the cost advantage shifts to aeration systems at water usages between 10,000 and 20,000 gpd.

From the beginning, it was recognized that decay of radon within the GAC bed would result in the growth of radon progeny. Figure 1 shows the uranium decay series, which includes radon and its daughter products or progeny. If these daughter products are retained in the GAC bed, the system will proceed toward radioactive equilibrium between radon and the progeny. In the case of beta and gamma emitting daughters Bi-214 and Pb-214, equilibrium with Rn-222 is attained within hours. Beta emitting Pb-210, because of its longer half-life, would take 22 years to reach fifty percent of equilibrium.

The health benefit that results from the removal of radon has overshadowed other radiological health considerations. It was recognized from the beginning that gamma radiation from a GAC filter could be a health hazard but perhaps this possibility was not fully appreciated.

Empirical Methods

Lacking evidence to the contrary, it was assumed in the early studies that radon progeny were totally retained on the GAC. This is an appropriately conservative worst case scenario when considering the health effects of gamma radiation from GAC beds. Later research has confirmed essentially total retention of radon daughters by GAC [8].

A typical domestic style water supply consists of a fiberglass or plastic tank 54 inches tall with an inside diameter of 10 inches and containing 2.5 cubic feet of GAC. The tank is operated in the down-flow mode and backwashing is done only when necessary to alleviate physical blockage of filter flow. Gamma profiles of a GAC tank in operation show that most of the radiation is emitted from the top several inches of the bed, as shown in Figure 2 [7]. Survey meter readings of the maximum gamma ray dose at several tank surfaces produced the following correlations between dose and influent radon concentration:

$$\text{Max mR/hr} = Rn_{inf}\ (\text{pCi/l}) / 17800 \quad (7)$$
$$\text{Max mR/hr} = Rn_{inf}\ (\text{pCi/l}) / 10360 \quad (8)$$
$$\text{Max mR/hr} = Rn_{inf}\ (\text{pCi/l}) / 8595 \quad (8)$$

While it is obvious that these relationships may differ greatly, a detailed discussion of the possible reasons for these differences is beyond the scope of this paper. However, it is important to note that this may be due largely to geometrical relationships between the radiation source and the survey meter detector. Large errors can result when the detector is in close proximity to the source.

There is a better way to measure the radiation environment around a GAC tank. A series of measurements made at various, often substantial, distances from the source provide the raw data for defining the radiation environment [8]. The gamma radiation dose at the source or at any distance is then proportional to the reciprocal of the distance squared.

To measure the gamma radiation field around GAC tanks, ten units were selected that had been in service between seven months and six years. Nine of these units are considered in this paper. The tenth unit, with an influent radon of 2620 pCi/l, produced so little radiation as to be virtually impossible to measure at a distance with acceptable accuracy. The tanks were disconnected and placed outside the installation building, and the gamma exposure field was measured by taking up to 400 readings within a two dimensional grid. The measurements of the distribution of gamma exposure rates surrounding the GAC vessel established isogamma lines from a maximum at the side of the vessel to the prevailing background of the installation area. This is illustrated in Figure 3.

To accompany the gamma data, accurate water use measurements were made by water meter. Radon measurements of raw and treated water were made 15 and 5 times respectively during the three week period preceding the gamma measurements. The existence of accurately established radon concentra-

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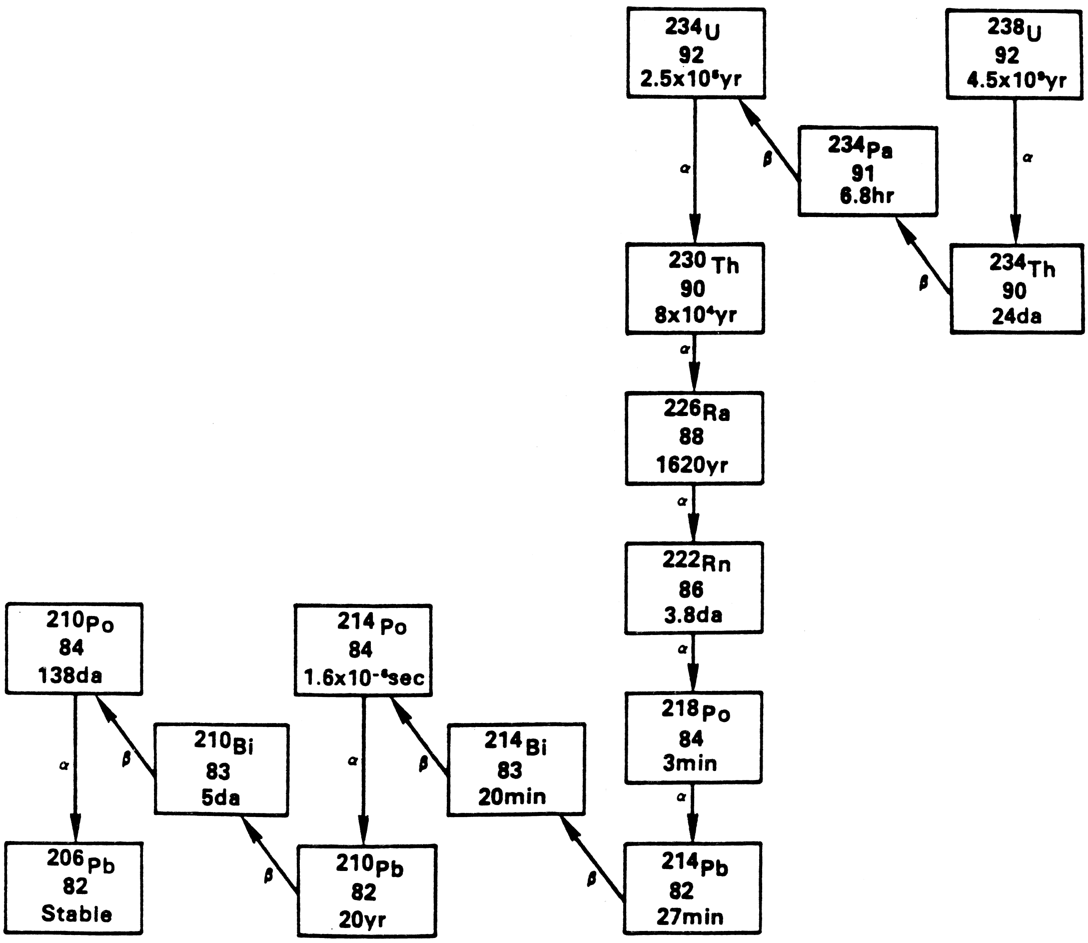
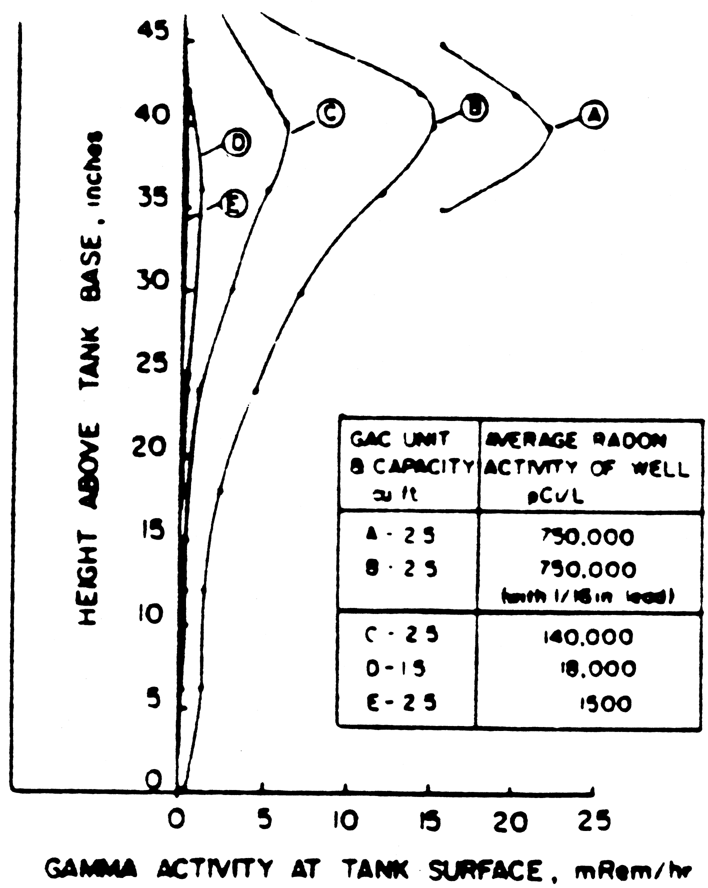


Figure 1
Principal Decay Mode & Half-Life



Gamma Activity Profiles for Several GAC Beds

FIGURE 2

(Taken From Ref. 7)

tions and water usages in addition to the gamma data make comparison between field measurements and theoretical calculations possible.

It should be noted that very large variations were recorded during the three week measurement period, in one case up to 900% difference between the maximum and minimum values for influent radon content. Radon concentrations that vary with time impact the representativeness of the information obtained from survey meter measurements or model calculations. Survey meter measurements are an "average" that is strongly biased toward the last several days because of radioactive decay considerations. Model calculations are based on an "average" radon content which is, one hopes, representative of the long term radon concentration.

Theoretical Calculations

In 1987, Keene and Rydell wrote a computer program called CARBDOSE that models radon removal by a domestic style GAC filter [11]. The program runs on microcomputers using the MS-DOS operating system (IBM-PC and compatibles) and calculates the following parameters:

1. The amount of radon (Rn-222) removed per day
2. The toal accumulated radon on the filter
3. The effluent radon concentration and estimated health risk
4. The probable exposure dose at 1 meter from the filter wall and at specified distances greater than 36 inches
5. The dose from an equivalent point source at any specified distance measured from the tank centerline
6. The distance at which the probable exposure dose from the GAC tank becomes less than the NCRP health guideline
7. The growth of Pb-210 on the filter for the number of years specified
8. The Pb-210 activity per gram wet carbon
9. The time when the carbon exceeds the 2000 pCi/gram waste disposal category

The current version of the program (version 2.0) utilizes the National Council on Radiation Protection Guideline for residential exposure [12]. This facilitates direct comparison with previously obtained field measurements of gamma exposure from GAC filters [8].

The computer program models a GAC filter by assigning all the radon removed to a cylindrical disk 10 inches in diameter and 5 inches thick. This is consistent with the observed gamma radiation associated with a thin layer at the top of the GAC bed [7]. The program treats the disk as a cylindrically corrected 24 cm x 24 cm x 13 cm array of 1 cm cubes. The dose originating from each cubic centimeter is calculated for the 72 gamma energies of Bi-214 and Pb-214 then corrected for self-absorption and the build-up factor associated with attenuation and scattering within the carbon/water volume distributed source. While decreasing the dose with distance from the source by the inverse square law, the model makes additional corrections for the absorption in air.

Generally the natural or ambient background radiation is a small, location-dependent, variable component of the total radiation exposure. For this reason, all exposure or dose values calculated by the program are exclusive of background. If background is known, it may be added to the calculated value to give a projected total exposure at a given distance. Our data shown below, plotted in terms of distance, indicate good agreement between projected total exposure and survey meter response.

In addition to the volume based dose, the model calculates an equivalent point source dose. The point source dose is corrected for distance and absorption in air only. The point source dose may be likened to assigning all radioactivity to the head of a pin located on the centerline of the GAC tank. While this is a conceptual abstraction of limited appeal when faced with the physical dimensions of the GAC filter, it serves a useful purpose in the internal calibration of the model and for possible extrapolation of the dose from small domestic style GAC filters to larger systems.

Results and Discussion

This paper is not an appropriate place for a protracted discussion of the question of absolute accuracy between survey meter results and those obtained through model calculation. Two survey meters with the capability of reading at the uR/hr level were used to measure gamma emission from the GAC tanks. The survey meters were laboratory calibrated with a Cs-137 standard just prior to the study. The model is calibrated to a theoretical Ra-226 standard.

Ideally, survey meters used for measuring the gamma doses resulting from radon removal by GAC would be calibrated with a Ra-226 standard. This is because equivalent meter response between unrelated radionuclides with differing gamma energies cannot be assured. However, Ra-226 standards are becoming quite rare as they have a disconcerting habit of leaking Rn-222 and radium. This in turn contaminates the surroundings with radioactive progeny.

The authors believe the measured values and the calculated values are in reasonably good agreement. Further, if one considers nothing more than the change in dosage that results from the natural variation in radon concentration with time, both types of values are sufficiently accurate to be useful in dealing with radiation health concerns.

One of the parameters that the model calculates is a distance, based on the probable dose from a volume distributed source, at which the dose becomes less than 0.058 mR/hr. The dose of 0.058 mR/hr is based on the National Council on Radiation Protection residential guideline value. The NCRP Bulletin [12] lists the following guidelines:

Occupational	5000 mR/year
Public	500 mR/year
Residential	170 mR/year

We have recalculated these as maximum permissible exposures for 8 hours per day. We think a person will be in his or her maximum proximity to a GAC filter for 8 hours or less per day. The recalculated guideline values are 1.172, 0.171 and 0.058 mR/hr respectively. For the residential situation, if you add the average natural background radiation (15 uR/hr) found for the field measurement sites in Maine, the result is the value of the 0.073 iso-gamma line shown in Figure 3. The above relationships are the basis for the comparison between measured data and the calculated data shown in Table 1 and various figures.

Figure 4 is a scatter diagram comparison of measured vs. volume source calculated distances for an 0.058 mR/hr exposure. The correlation coefficient is 0.985. The equation of the regression line is, Y = -21.136 + 1.167 * X. Similar values are found for the point source data. We do not attach any further significance to this other than what appears to be an indication of good agreement between measured and calculated values.

The values shown in Table 1 without site numbers are artificial input values used to supplement actual values and demonstrate that the model response is uniform even at the low end of the scale. This is illustrated in Figure 5.

The lines designated Calibration A & B in the legend of Figure 6 are plots of calculated data sets that do not appear to be a good fit with the survey meter measured values. Calibration A is corrected to the theoretical Ra-226 standard and has a carbon/water effective density of 1.1 The theoretical Ra standard is the dose received from a Ra-226 source of known specific activity encapsulated in 0.5 mm of platinum (density 21.4) after correction for the absorption of the platinum. Calibration B is for a carbon/water effective density of 1.1 but does not include correction to the radium standard. Best agreement between measured and calculated values are for the lines designated Model (Pt) and Model (Vol) both of which are corrected to the radium standard and have a carbon/water effective density of 1.0. We have observed that backwashing domestic GAC filters at all but the lowest flow rates results in fluidization of the bed, hence the carbon/water effective density is quite low, but a density of 1.0 is the absolute minimum of the possible range.

As we indicated earlier in this discussion, the question of absolute accuracy is moot. However, the fact that the best agreement between methods is obtained with a somewhat suspect density suggests that there is a small calibration error in one or both methods.

The calculated point source dose from a given amount of radon is consistently higher than the volume

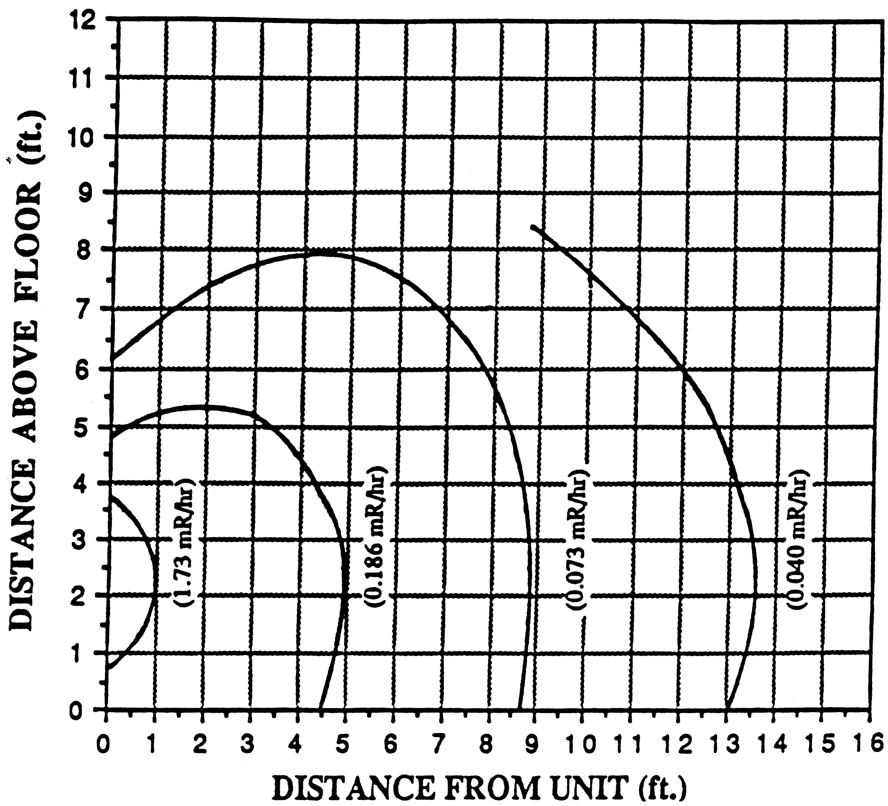


Figure 3
Gamma Exposure Rate Field Surrounding GAC Unit at Site 6
95,550 pCi/l Rn-222
Max. Gamma: 9.000 mR/hr
(Taken from Ref. 8)

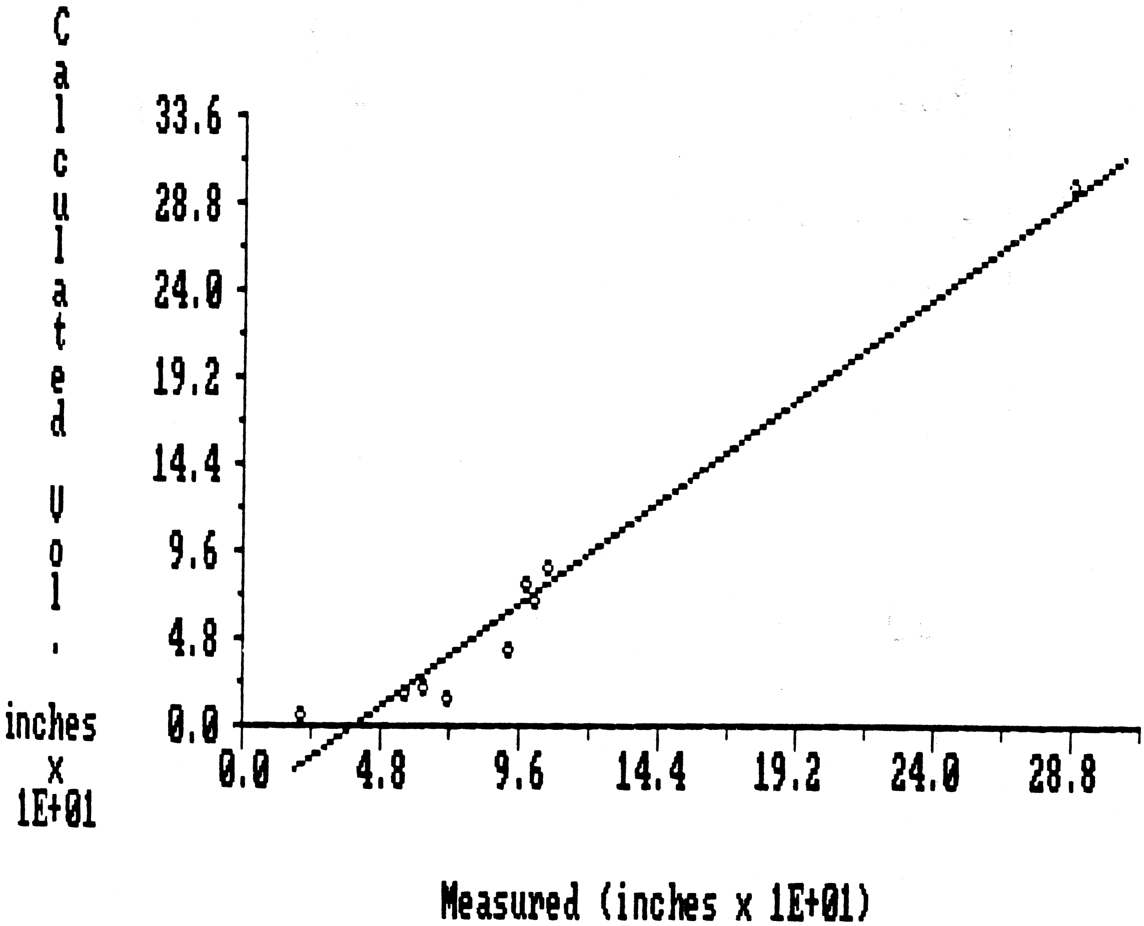


Figure 4
Scatter Diagram of Measured vs. Calculated Distances
for 0.058 mR/hr

Site No.	Radon pCi/1	Percent Radon Removal	Daily Usage 1/day	Daily Slug uCi/day	Field Measured (in.)	Model Calc. Vol. Sor. (in.)	Model Calc. Pt. Sor. (in.)	Pb-210 >2000 pCi/gram (years)
*				0.099		3.900	4.230	
*				0.198		5.500	5.980	
*				0.495		8.700	9.450	
*				0.990		12.300	13.370	
*				1.980		17.500	18.910	
*				3.960		24.700	26.740	
2	16540	99.640	329.0	5.422	20.0	28.900	31.290	17.9
5	53700	91.710	209.0	10.293	71.0	39.800	43.110	8.2
4	40800	83.180	362.0	12.285	56.0	43.500	47.100	6.7
3	34600	99.570	403.0	13.884	63.0	46.200	50.070	5.9
10	69300	99.700	418.0	28.880	92.0	66.700	72.220	2.7
7	107200	99.020	532.0	56.472	101.0	93.200	100.990	1.4
8	125600	99.430	553.0	69.061	98.0	103.100	111.680	1.1
6	95550	91.660	939.0	82.239	106.0	112.500	121.870	1.0
9	758800	99.380	886.0	668.129	288.0	320.600	347.360	0.2

* Artificial Values

Table 1
Measured and Calculated Data for Domestic GAC Filters

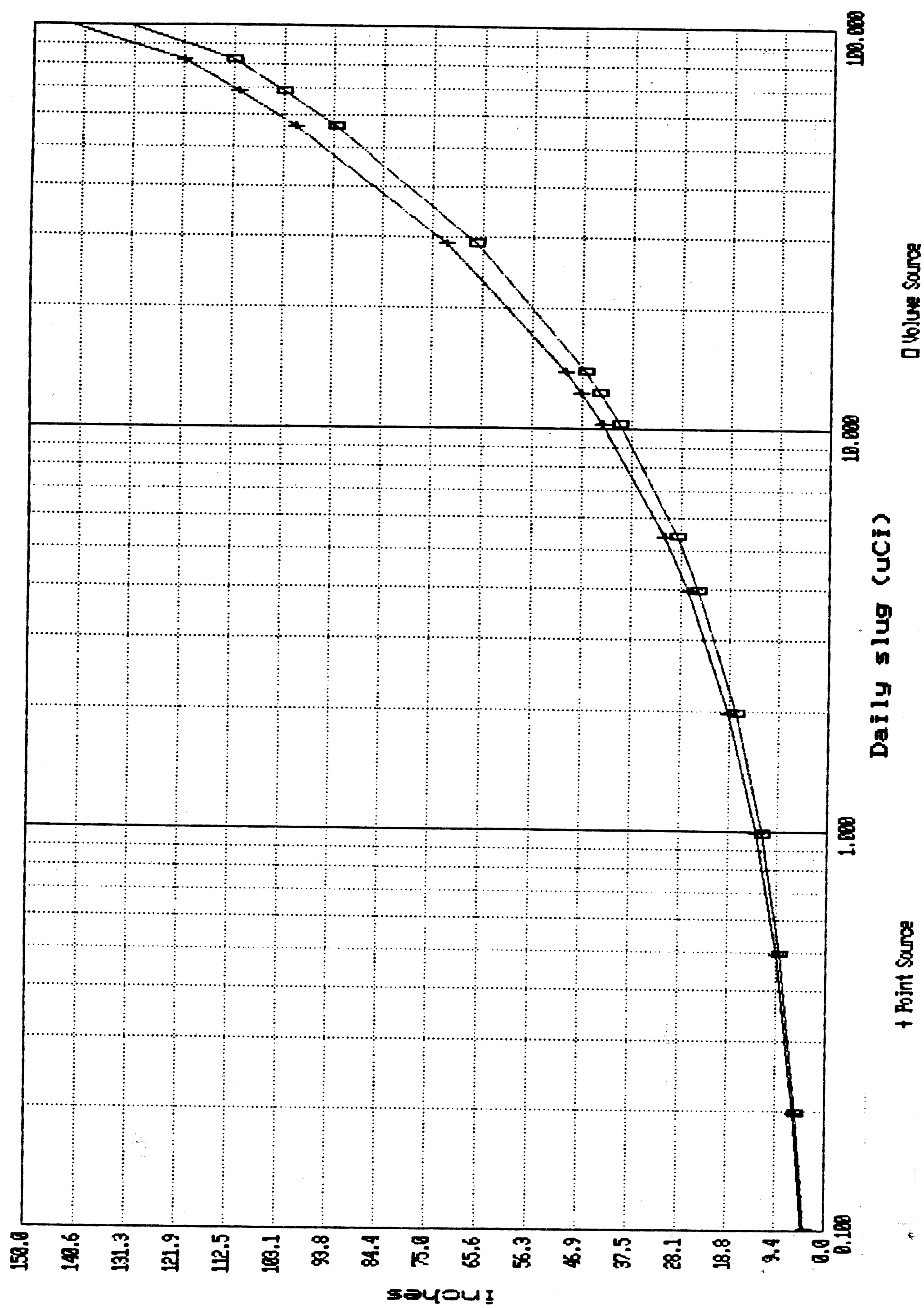


Figure 5
CARBDOSE Internal Model Behavior
Point Source vs. Volume Source
for 0.058 mR/hr

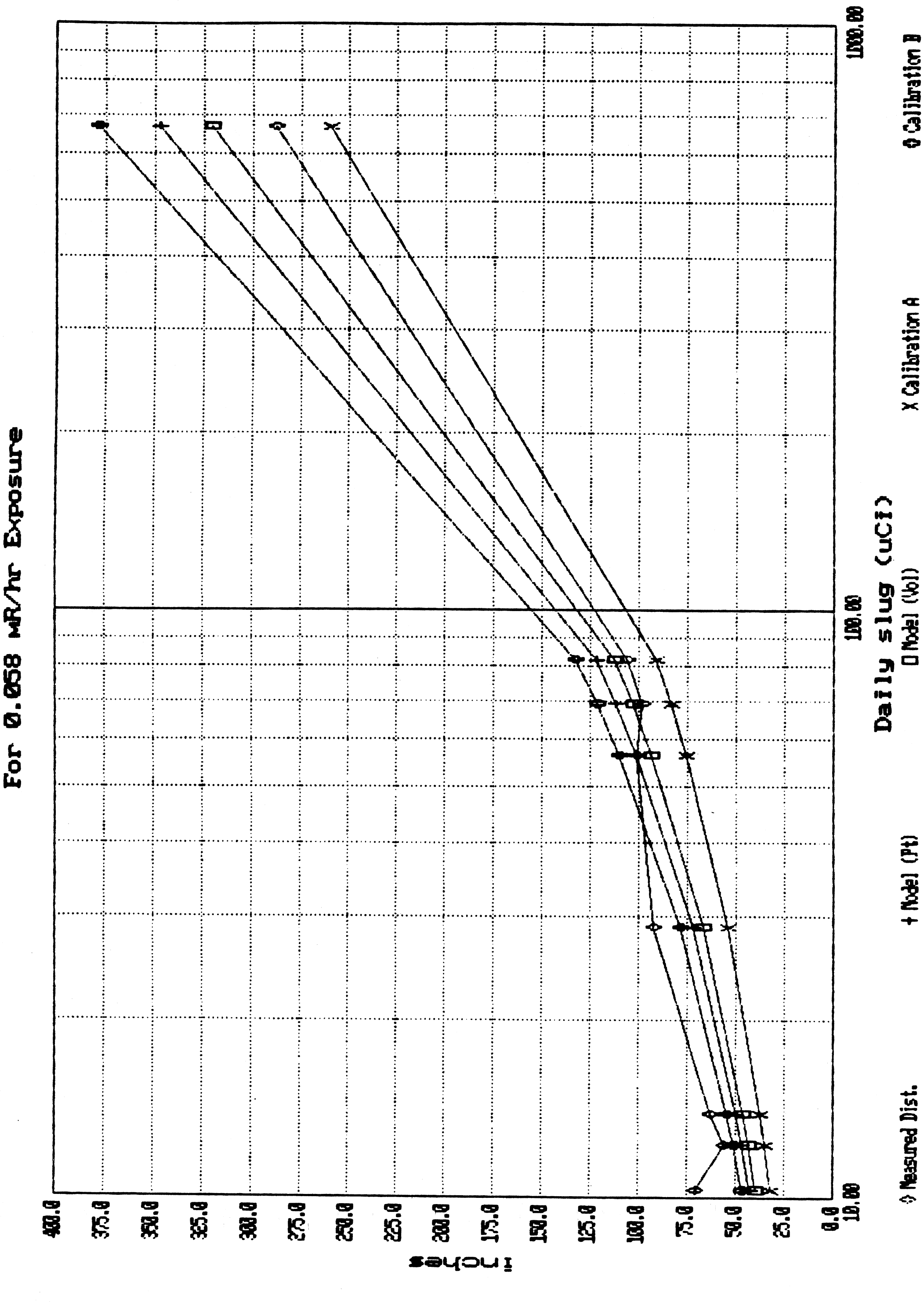


Figure 6
Comparison of Measured and Calculated Distances
for Various Model Conditions

source dose. The data plotted in Figures 6 & 8, where the dose is held constant, show point source values attaining this dose at greater distances than the corresponding volume source values. Point source doses and/or distances are higher because the model does not correct for the attenuation of radiation by the carbon and water in the filter.

Use of the radon content, as in the empirical relationships and Figure 7, erroneously shows radiation exposure as an exclusive function of radon content. To overcome this we report data as daily slugs, a convenient unit for working with radiation from GAC filters. We define the daily slug as the product of the radon content times radon removal efficiency times water usage per day. When comparing Figure 7 and Figure 8, it is seen that plotting data in terms of daily slugs gives a smoother, more easily understood graph. More importantly, the daily slug is really what the survey meter responds to and the model calculations are based on.

The authors feel that the volume source distance line shown in Figure 8 is the best fit with the survey meter measured distances. This is a satisfying conclusion to reach, as the volume source model is the one that most completely addresses the geometric factors and the build-up and self adsorption factors that contribute to the total gamma radiation dose.

Lead 210 Accumulation

Lead 210 and its daughters are beta and alpha emitters without major associated gamma activity. Hence, the risk from them is largely one of contact, particularly through ingestion or inhalation.

As long as the GAC filter operates it will continue to accumulate Pb-210. The half-line of Pb 210 is 22 years, therefore its activity will continue to grow until radioactive equilibrium with radon is achieved in about 100 years. Long before equilibrium is attaned, Pb-210 and its progeny Bi-210 and Po-210 have the potential to become problems should the GAC need regeneration or disposal. In addition to being an alpha emitting radionuclide, Po-210 is easily volatilized.

The Environmental Protection Agency is in the process of proposing a set of guidelines for the disposal of naturally occurring radionuclides generated by water treatment plants. One of the EPA guidelines applicable to our discussion is that of solid wastes containing more than 2,000 pCi/gram (dry weight) of natural radioactivity. These should be disposed of in a low-level radioactive waste disposal facility operated under the provisions of the Atomic Energy Act. At present, there are only a few disposal sites in the nation that meet these criteria.

The computer program calculates the time, for the conditions given, when the Pb-210 activity exceeds 2,000 pCi/gram (wet carbon basis). The times for the field measurement sites are given in Table 1, and over half are less than five years. The time when the carbon radioactivity, on a dry weight basis, would exceed 2,000 pCi/g is considerably shorter.

Conclusions

Early enthusiasm for GAC as the radon treatment medium of choice for very small systems has diminished in consideration of the secondary radiation problems it presents. In the authors' view, GAC remains a viable treatment method for radon only at the low end of the radon concentration range. In the domestic water supply situation, this is 5000 pCi/l or less.

The initial cost advantages of the GAC system may be overridden by the potential costs of dealing with problems of gamma radiation shielding or the need to regenerate or dispose of radioactive spent carbon should the system become plugged or otherwise fail.

Calculations of gamma dose via model allow design estimates for domestic systems to be made with reasonable accuracy. Using the model's point source dose feature should enable water system designers to estimate radiation exposures for larger units, since the point source calculation is independent of the shape or size of the source, and does not consider self-absorption within the source.

On site verification of gamma dose by a recently calibrated survey meter after several weeks of filter operation is good engineering practice, and, in our opinion, should be done to check the accuracy of the model calculation.

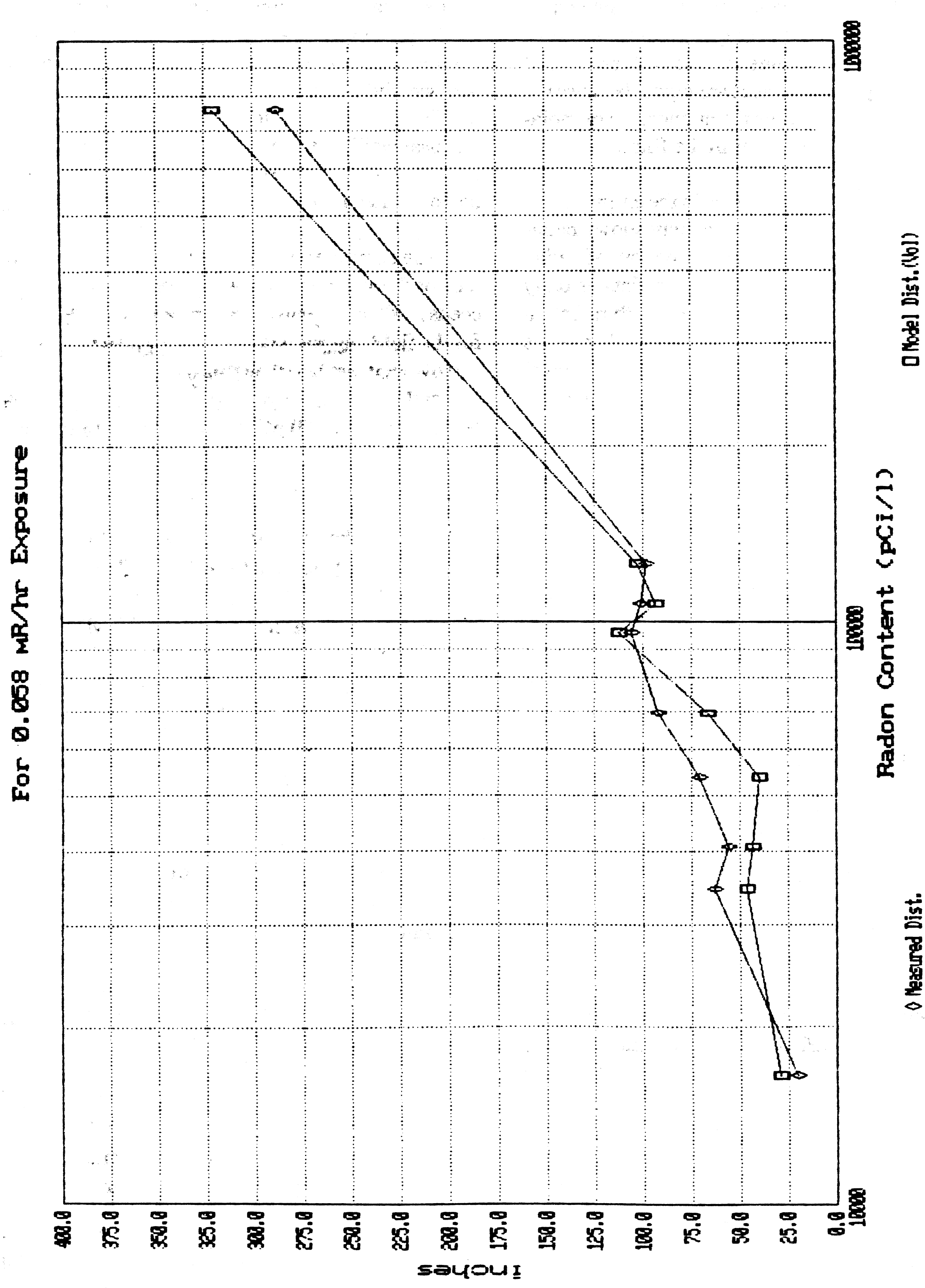


Figure 7
Measured and Calculated Distances
as a Function of Radon Content

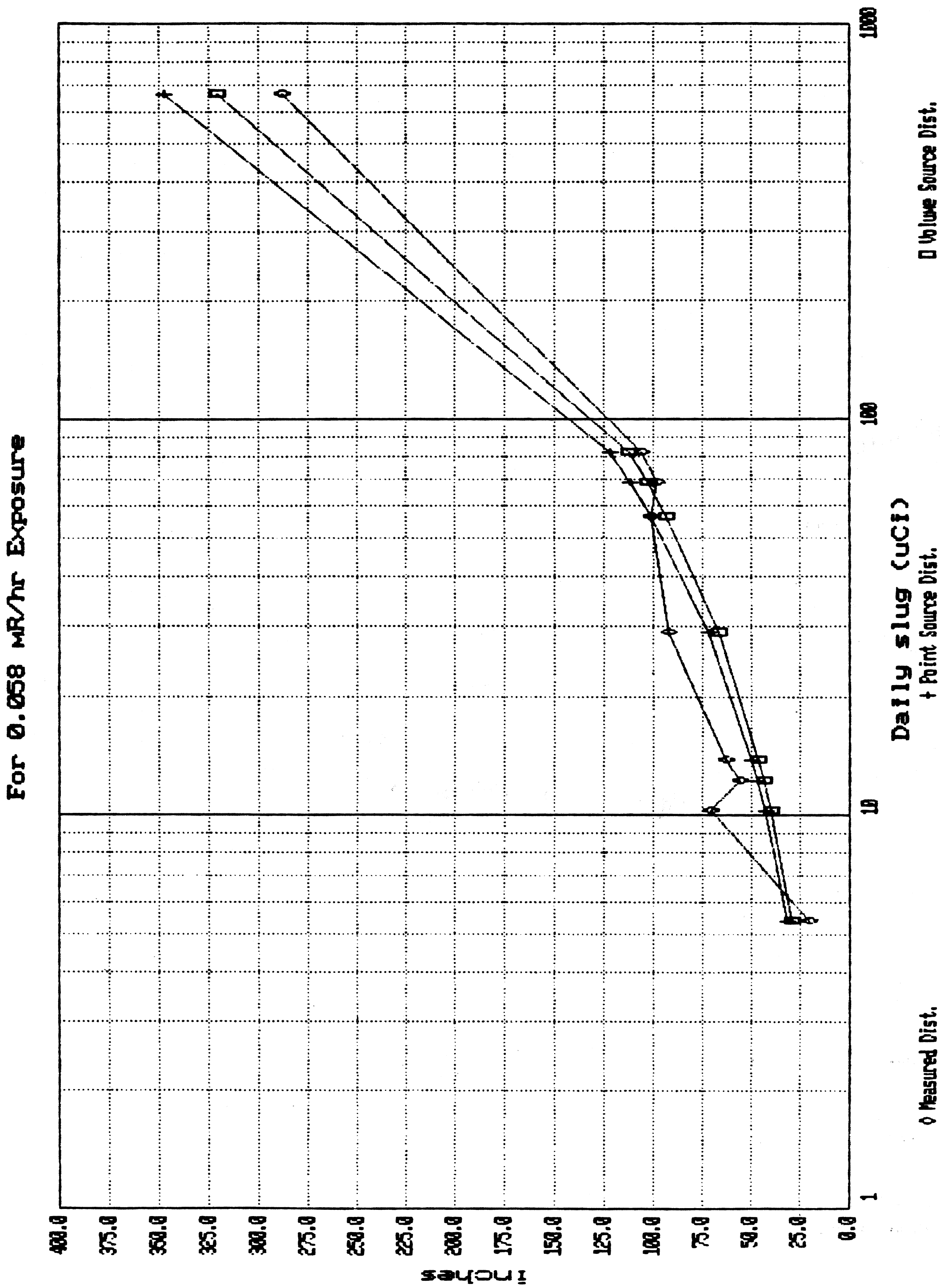


Figure 8
Comparison of Point Source and Volume Source
with Measured Values

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This paper, while not presented before a meeting of the association, was offered for publication by the authors. Because of the current concern about Radon in drinking water and its public health implications, it was deemed important to bring this information to Journal readers in a timely manner.
Editor

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12. National Council on Radiation Protection Report No. 39, 1971.