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POTABLE WATER AS A SOURCE OF AIRBORNE RADON-222 IN U.S. DWELLINGS: A REVIEW AND ASSESSMENT

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ABSTRACT

Using a long-term-average, single-cell model and available data for U.S. housing, the concentration of radon in indoor air due to the use of potable water is assessed. The ratio of the airborne radon concentration to the concentration in water is represented by a lognormal distribution with geometric mean and geometric standard deviation of 0.65 x 10^{-4} and 2.88, respectively, in fair agreement with the previously reported results of direct measurements of the ratio in 13 houses. By combining this result with data on radon concentrations in U.S. water supplies, potable water is estimated to contribute an average of 24, 1.3, and 0.1 Bq m^{-3} to the airborne radon concentration in residences served by private wells, public ground water, and surface water supplies, respectively.

Keywords: air-exchange rate, building volume, indoor air quality, pollutant sources, radon, residential buildings, water, water-use rate

INTRODUCTION

In recent years, exposure in buildings to the radioactive progeny of radon-222 has gained prominence as an element of the natural radiation environment. In fact, as a result of numerous studies, such exposure is now estimated to account for nearly 50% of the effective-dose-equivalent to the public in countries with temperate climates (UN82). Consequently, vigorous research activity has investigated many aspects of the indoor radon problem: determining indoor concentrations; examining the effects of various controlling factors, such as ventilation rate; characterizing the source materials and transport processes by which radon enters houses; and studying the behavior of radon progeny in indoor air. (See, for example, Ne83.) These investigations have revealed the importance of variability in source strengths in determining the broad range of indoor concentrations that has been observed. The predominant source of radon in U.S. houses generally appears to be the soil adjacent to the foundation (Ne84a). The other major sources -- potable water and building materials -- often contribute significantly to indoor concentrations, and either may be the dominant source in some cases.

High radon concentrations in potable water were first observed in Maine in the late 1950's (Sm61). Initially, concern about radiation hygiene as a consequence of these observations focused on ingestion of radon-rich water, and researchers identified the stomach as the organ receiving the greatest dose (Tu61, Do64, Hu65, He66, Su72). Later, Gesell and Prichard speculated that the inhalation exposure to radon progeny arising from the release of radon from domestic water uses may be of greater significance than the ingestion exposure (Ge75). More recent studies, comparing the expected lung and stomach doses resulting from radon-rich water, have concluded that the former is three to twelve times the latter (Du76, Ka80, Pr81).

Several studies in the past decade have investigated aspects of potable water as a source of airborne radon indoors. Key efforts in the United States include these: the development of an inexpensive technique for measuring radon in water (Pr77); studies of the transfer of radon from water to air during different household uses (Ge80, Pa79, He82); measurements of the indoor airborne radon concentration resulting from water use in different residences (Ge80, He82); estimation of the population dose resulting from radon in potable water supplies in Houston, Texas (Pr81); and surveys of radon concentrations in groundwater supplies (Pr83, Ho83). In addition, numerous investigations have been conducted in Finland, where groundwater radon concentrations are particularly high (As79, As80, Ca80, Ka80).

The purpose of this paper is to review the state of knowledge of domestic water as a source of indoor airborne radon, focusing on the assessment of indoor radon

concentrations that result from water use. Available data on residence volumes, air-exchange rates, in-house water-use rates and water-to-air transfer coefficients are combined to determine the average incremental indoor radon concentration in air that can be expected to result from a specified concentration of radon in water. The distribution of radon concentrations in U.S. water supplies is discussed, and the contribution of potable water to indoor radon concentrations is estimated.

PREDICTING THE INDOOR RADON CONCENTRATION RESULTING FROM WATER USE

Long-Term-Average Single-Cell Model

From conservation of mass, the average indoor radon concentration resulting solely from water use can be estimated from the following equation (see appendix for derivation):

$$C_a = \frac{C_w W e}{V \lambda} \tag{1}$$

where

C_w is the radon concentration in water entering the residence (Bq m^{-3}),
W is the water-use rate per resident (m^3 $person^{-1}$ hr^{-1}),
e is the use-weighted average transfer efficiency of radon from water to air (dimensionless),
V is the volume per resident of the dwelling (m^3 $person^{-1}$), and
λ is the air-exchange rate of the residence (hr^{-1}), assumed to be much greater than the decay constant for radon (0.0076 hr^{-1}).

An important question in assessing the impact of radon in water on exposure to radiation is this: given a concentration of radon in water, C_w, what is the expected increase in the concentration in air, C_a, that results from water use? The answer to this question can be expressed in terms of a transfer factor, f:

$$C_a = f C_w , \tag{2}$$

where

$$f = \frac{We}{V\lambda} . \tag{3}$$

There are at least two distinct approaches to determining f. The first, which we will use in this paper, evaluates e, W, V and λ from an examination of literature, then calculates the distribution of f by combining these parameters mathematically. The second approach determines C_a and C_w experimentally in a sample of houses and then evaluates f directly from equation (2). In using the former approach, previous studies have specified one or more typical values of V and λ, combined them with limited data on W and e,and calculated f (Pa79, Ge80, Be84). They have concluded that the most typical value of f is in the vicinity of 10^{-4} and that the value for a specific residence can be an

order of magnitude smaller or larger.

Two studies in the United States have evaluated f using the experimental approach. In the first of these, radon was monitored continuously for several days in each of four residences (Ge80). The difference between average radon concentrations for the periods 0800-2400 and 0000-0800 was assumed to be due to radon liberated by water use. In the second study, identifiable peaks in indoor radon concentration in 13 houses were used to assess the contribution to indoor concentration from dishwashing, showering and laundry (He82). The contribution of other water uses -- toilets, baths and sinks -- was based on estimated transfer coefficients and water-use rates. The results of these studies are discussed in a later section.

The results of measurements in seventeen houses in two small regions is not sufficient to precisely estimate the distribution of f in the U.S. housing stock. Yet, determining f in a large number of houses using experimental techniques such as these poses substantial difficulties. The radon contributed by water use must be separated from that due to other sources. Thus, the small fraction of houses in which water is the predominant source are best suited as study sites. Even in these cases, contributions from other sources, particularly soil and outdoor air, must be accounted for -- a necessity that is complicated by the potential for large temporal variability in entry rates from these sources (see, *e.g.*, Na85). The approach presented here, mathematically combining data on the contributing factors to determine the distribution of f, constitutes a more feasible alternative for obtaining statistically-robust results. An alternative experimental approach is to measure W, e, V and λ in a randomly selected set of houses and evaluate f for each house from equation (3). Although this approach appears to offer advantages over the other experimental technique, particularly in avoiding interference from radon sources other than water, it has not yet been used.

Water-Use Rates

The results from several studies giving residential in-house water-use rates per occupant for specific functions are listed in Table 1. Four of these studies report results from 29 individual residences (Co74, La74, Be75, EPA78). Two additional studies, not listed in Table 1, give the total in-house consumption rate for 18 homes in Louisville, Kentucky (An67) and 43 homes in Wheatland and Laramie, Wyoming (Ba79). Total in-house use rates per person for these 90 residences are plotted as a cumulative probability distribution in Figure 1. The data are well-fitted by a lognormal distribution with a geometric mean of 7.9×10^{-3} m^3 $person^{-1}$ hr^{-1} (0.189 m^3 $person^{-1}$ day^{-1}) and a geometric standard deviation of 1.57. The null hypothesis that the parent distribution is lognormal cannot be

rejected, even at a significance level of 0.2, according to the Kolmogorov-Smirnov test (Af72). Three previous studies of indoor radon from potable water have cited water-use rates of 0.152 (He82), 0.223 (Ge80) and 0.262 (Pa79, Ho84a) m^3 $person^{-1}$ day^{-1}; each of these values is within one geometric standard deviation of the geometric mean determined here.

Transfer Efficiency

Radon is relatively insoluble in water. Boyle measured the solubility coefficient as a function of temperature and found values of 0.51, 0.25 and 0.16 at 0.0, 20.0, and 39.1 ^{o}C, respectively (Bo11). As an example of the significance of these data, consider a toilet tank holding 0.02 m^3 of water at 20 ^{o}C in a closed bathroom of 15 m^3. At equilibrium, assuming the initial concentration in air is zero, 99.97% of the radon originally in the water has been released. Measurements have shown that the actual fraction released in residences is generally much smaller, indicating that equilibrium is not attained. A complete theoretical analysis of the transfer of radon from water to air in households has not been undertaken; instead, researchers have measured the fraction released experimentally.

Transfer coefficients have been determined for major household water uses in three studies. In each case, concentrations of radon in the inlet and effluent streams were measured. In one study transfer coefficients for some uses were estimated from laboratory investigations of the dependence of the liberated fraction on agitation and on the surface-to-volume ratio of the water reservoir (He82). The results from these studies are summarized in Table 2, which shows good agreement among them.

To determine a use-weighted mean transfer efficiency we have applied mean coefficients by use (the last column in Table 2) to average water use by function for the 21 houses for which use by function was comprehensively monitored (La74, Be75, and EPA78 in Table 1). The results are plotted as a cumulative frequency distribution in Figure 2, which shows a geometric mean of 0.55 and a geometric standard deviation of 1.12. The range for the 21 houses considered was small, 0.44 - 0.68, leading to the conclusion, as discussed later, that variability in the transfer efficiency is likely to be a small factor in the overall variability of f.

House Volume

In a study conducted for the U.S. Department of Energy, the floor area of the heated portion of 6051 randomly selected residences was measured (RECS82). The report tabulates the number of residences in each of seven building size classes for six categories

corresponding to the number of household members. After assuming a fixed ceiling height of 2.4 m, we applied a least-squares analysis to the cumulative frequency plots of these volume data to derive lognormal statistics for volume per resident as a function of the number of residents per household. (See Figure 3.) These statistics were then combined mathematically, with weighting factors proportional to the number of people in each class, to obtain 99 m^3 $person^{-1}$ and 1.90 as the geometric mean and geometric standard deviation, respectively, for the U.S. population. This distribution is plotted as the bold line in Figure 3 and is seen to agree well with the points enclosed by circles which were determined by graphically aggregating the results from the other six curves.

Air-Exchange Rate

Two studies have been published that give air-exchange rate data for a large number of U.S. residences. In the first of these, 1048 tracer-gas decay measurements, each giving the air-exchange rate over a few hours, were made in 266 dwellings occupied by low-income families and located in 14 cities spanning the major U.S. climactic zones (Gr81). The individual-measurement results are plotted as cumulative frequency curve "a" in Figure 4 and are fit by a lognormal distribution whose geometric mean and geometric standard deviation are 0.9 hr^{-1} and 2.13, respectively.

The second study evaluated average infiltration rates for the November-March heating season in 312 residences (Gr83). In contrast to the first study in which the median house age was 45 years, the houses in this investigation tended to be fairly new, with a median age of less than ten years. Furthermore, these houses did not represent the distribution across U.S. climate zones: most were located in Washington, California, Colorado, New York and Ontario, Canada. The cumulative probability distribution of infiltration rates for these houses is plotted as curve "b" in Figure 4. The Kolmogorov-Smirnov test for goodness-of-fit indicates that the hypothesis that the parent distribution is lognormal can be rejected at a significance level of 0.05, although not at a significance level of 0.01.

As these two studies concentrated respectively on low-income houses, which are probably leakier than average, and on modern houses, which are probably tighter than average, we expect that the distribution for all U.S. housing lies between the two. We have estimated the true distribution by aggregating the two distributions weighted proportionally by the number of houses. The resulting geometric mean is 0.68 hr^{-1} and the geometric standard deviation is 2.01,

A caution must be noted in using this result: the measurements apply for the most part only to infiltration and only during the heating season. In many cases air-exchange

rates at other times of the year are higher due to windows being open. If windows and doors are closed, however, the air-exchange rate may be lower because the milder weather conditions lead to reduced driving forces for infiltration.

The Distribution of f

A variable derived as the sum of independent, normally-distributed parameters is itself normally distributed with a mean value given by the sum of the means and a variance given by the sum of the variances (Hi80). Similarly, a variable that is the product of independent, lognormally distributed parameters is itself lognormally distributed. Thus, given the lognormal statistics for W, e, V and λ, as summarized in Table 3, the lognormal statistics for f can be determined as follows:

$$GM_f = \frac{GM_W \, GM_e}{GM_V \, GM_\lambda} \tag{4}$$

$$GSD_f = \exp\left[((ln(GSD_W))^2 + (ln(GSD_e))^2 + (ln(GSD_V))^2 + (ln(GSD_\lambda))^2\right]^{1/2*} \tag{5}$$

In these expressions GM_i represents a geometric mean, GSD_i represents a geometric standard deviation and the subscripts designate a parameter. Using this approach to derive the distribution of the overall air-to-water ratio yields a geometric mean for f of 0.65 x 10^{-4} with a geometric standard deviation of 2.88.

There are two methods of comparing this result with those of other studies. First, we can calculate the arithmetic mean of f from the following equation:

$$AM = GM \exp\left[(ln(GSD))^2/2\right]^{**} \tag{6}$$

The resulting value, 1.14 x 10^{-4}, agrees well with the typically cited value of 1 x 10^{-4}. The second approach compares this distribution with that resulting from direct measurements of C_a/C_w. For this comparison we have constructed a plot, shown in Figure 5, of

* The validity of assuming that the variables on the righthand side of this equation are independent is examined in a subsequent section.

** The minimum variance unbiased estimator of AM is given by a somewhat more complex function which depends on the number of samples as well as the geometric mean and geometric standard deviation (Ai57). For a large number of samples the expression converges to equation (6). Taking n=21 for the current case (corresponding to the number of determinations of e), the arithmetic mean determined by the more complex formula is 1.10 x 10^{-4}.

the cumulative frequency distribution of derived from data for 13 houses in which C_a/C_w was directly measured and in which the radon concentration in water exceeded 40,000 Bq m^{-3} (Ge80, He82). In four other houses in which such measurements were made, the waterborne radon concentrations were so small that the expected contributions to radon in air were small compared even to outdoor concentrations and therefore extremely difficult to measure. Considering the widely different means of determining f, the agreement between our computed distribution and the results of direct measurements is reasonably good: twelve of the thirteen measurements lie between the 5% and 95% contours of the computed distribution.

The fifth column in Table 3 (labelled % var.) indicates the relative contributions of the various factors to the range of the distribution of f. These data indicate that differences among houses in volume per resident and air-exchange rate are more important than differences in water-use rate and transfer efficiency in accounting for the differences in f among the housing stock.

In interpreting the results of this analysis, one must recognize the distinction between the variance associated with the distribution of a parameter and the uncertainty in the estimate of its mean. The former, reflected by the column labeled "GSD" in Table 3, provides information on the range of actual values in the parent distribution. The latter, reflected by the column labeled "GSE", indicates the degree to which the data are adequate in number to determine the mean of the parent distribution. Thus, a GSD 2.88 for f suggests that the value of f for 68% of the housing stock is contained within the range (0.23 - 1.87) x 10^{-4} -- *i.e.*, within a factor of 2.88 of the geometric mean. On the other hand, a GSE of 1.063 for f indicates that the 90% confidence limits on its geometric mean (assuming the sampling to be representative) are 0.58 x 10^{-4} and 0.73 x 10^{-4}. The data in the GSE" column also indicate that the water-use rate data contribute the greatest amount of the four factors to the uncertainty in GM_f, suggesting that additional experimental work to improve the estimate of f be directed at assessing W for a larger number of households.

From these results we can also determine the number of households needed to estimate GM_f by direct experimental measurement with a precision comparable to that of the present analysis. That number is given by

$$N = \left[ln(GSD_f)/ln(GSE_f) \right]^2 = 300. \qquad (7)$$

Thus, f would have to be measured in 300 residences to improve the statistical basis for estimating the parameters of its distribution. This analysis does not consider how well

the data represent the U.S. housing stock. Because the sampling for each parameter except V cannot be considered random, the uncertainty in our estimate of the geometric mean of f is larger than that indicated by the *GSE*. However, an approach to improving the estimate of the distribution of f that uses direct experimental measurements would also experience difficulties in selecting a representative sample.

RADON CONCENTRATIONS IN POTABLE WATER SUPPLIES

Radon concentrations in water have been observed to range over an extremely large range, from effectively zero to more than 10^6 Bq m^{-3}. Surface waters, which serve 49.5% of the U.S. population (So83), have the lowest concentrations. Private wells, serving 18.3% of the population, generally have the highest concentrations, whereas public groundwater supplies, which serve the remaining 32.2%, have intermediate concentrations. In assessing the data on radon in water supplies, we consider these three sources separately.

Considerable work has recently been undertaken by the U.S. Environmental Protection Agency (EPA) to measure radon concentrations in public groundwater supplied in the United States. (This research is summarized in Ho83.) Using these data, along with other results from literature, we have computed lognormal statistics for the 41 states for which data exist (See Table 4). In computing these statistics, the logarithm of each concentration measurement was weighted by the inverse of the logarithm of the variance due to counting uncertainty. The analysis was complicated by two features of the data: 1) due to counting uncertainty, some of the measurement results were negative: and 2) in early reports, measurements below the detection limit of approximately 600 Bq m^{-3} were reported as "not detectable". Consequently, the parameters of the distribution could not be determined from the first and second moments of the entire set of log-transformed data. Instead, we estimated the geometric mean as the median of the entire set of data and determined the geometric standard deviation by calculating the second moment (about the median) of the measurements whose value exceeded the median.

To determine the population-weighted average for the country, the statistics for each state were combined with weights proportional to the population using public groundwater (So83). The result was a geometric mean of 5.2×10^3 Bq m^{-3} and a geometric standard deviation of 3.53. In addition to the caveat noted above, a few other cautions should be noted in interpreting this result. First, it is slightly positively biased by the fact that sampling was done at distribution sites, rather than at household taps. A more important concern is potential sampling bias. In the entire EPA study, the supplies that were to be sampled were chosen by state officials with no randomness criteria specified by EPA. Furthermore, for most of the study, sampling was limited to supplies serving 1000 or more people. While 86% of the U.S. population that is served by public groundwater is accounted for within this criteria, limited data show concentrations as much as an order of magnitude greater in smaller supplies (He85).

Higher concentrations have also been observed in private wells, although the data are quite limited. For the present work, the assessment was based on 44 measurements

from nine states (EPA79a, EPA79b, EPA79c, EPA80). The geometric mean and geometric standard deviation using an equal-weight calculation were found to be 36 x 10^3 Bq m $^{-3}$ and 6.5 respectively. The dominant uncertainty in these results is due to unrepresentative sampling; hence a more rigorous analysis of the data is unwarranted.

Information on radon concentrations in surface water supplies was derived from this same set of reports. Using a linear regression to the cumulative frequency plot of the 13 of 38 measurements which exceeded the detection limit of 0.6 x 10^3 Bq m^{-3}, the geometric mean and geometric standard deviation were estimated to be 0.3 x 10^3 Bq m^{-3} and 5.0, respectively.

Geometric mean radon-222 concentrations for these three types of water supply were recently reported by Hess *et al.* (He85). For private wells and public groundwater supplies, the geometric means for the entire country agree within 10% of those reported here. Values for public groundwater supplies in individual states differ markedly however. These differences appear to be due, in part to: (1) the number of samples reported in the present work for the individual states are one per water supply system, while Hess *et al.* treat duplicate samples from a single water supply as two separate samples; (2) for data falling below the minimum detection limit, Hess *et al.* assigned an arbitrarily small value ($\sim$4 Bq m^{-3}) for those data reported as zero or with negative values; and/or (3) for some states Hess *et al.* incorporate in their analysis early data not included in the subsequent published EPA reports. In some cases these early data may have been collected as part of an effort to look for high concentrations of radon in water supplies rather than to ascertain the actual concentration distribution (Ho85). The aggregate geometric mean reported by Hess *et al.* for surface water supplies is almost an order of magnitude smaller than the value we have determined. For either case, however, the conclusion remains that compared to other sources, surface water supplies are a negligible factor in contributing to indoor radon.

By combining the distributions of C_w with the distribution of f, one obtains an estimate of the parameters of the distribution of C_a, the airborne radon concentration in residences attributable to water. Following the approach used in deriving equations (4) - (6), the distribution parameters were determined for the three types of water supply, as shown in Table 5. Also shown in this table are the fraction of houses in which the indoor radon concentration due to use of potable water exceeds two benchmark values. The lower value, 9.3 Bq m^{-3}, corresponds to the typical concentration entering residences from the outdoor air (Ge83). The higher value, 33 Bq m^{-3}, corresponds to the estimated geometric mean radon concentration in U.S. residences (Ne84b). In only a small fraction of houses served by public groundwater supplies is water a major source of indoor radon.

On the other hand, the limited data on radon concentrations in private wells suggests that a substantial fraction of these may constitute important sources of indoor radon for homes utilizing private well water.

These results also indicate that public ground water supplies cause about 0.8% of the radon progeny exposure for the total U.S. population; the total exposure is estimated to cause an average individual lifetime risk of lung cancer of 0.15% (Jac84, Jam84, Ne84b). Thus the *average* lifetime risk due to radon from public water supplies is in the vicinity of 10^{-5}, equal to the *limiting* risk commonly used by governmental agencies for regulation of water and airborne environmental agents. Considering the broad distribution of C_a, a significant number of people are exposed to airborne radon from public water supplies at levels corresponding to risks of 10^{-4} to 10^{-3} or even higher. Given this observation, it is ironic that even if these supplies were controlled to the extent that they contributed nothing to indoor radon concentrations, the average public risk from exposure to airborne radon progeny would be essentially unchanged. These seemingly inconsistent features arise because, although 99% of indoor radon originates in sources other than public groundwater supplies, estimates suggest that the health risks associated with typical indoor radon levels are unusually high relative to those associated with other environmental pollutants.

LIMITATIONS OF THE CURRENT ANALYSIS

There are several respects in which the analysis presented here may be criticized. These weaknesses are briefly discussed below, focusing on the estimation of the parameters of the distribution of f. Some could be diminished by collecting additional data; others are intrinsic in the approach and would require an unjustifiable level of effort to rectify; none, in our opinion, are likely to greatly affect the resulting estimates.

Use of the Lognornal Distribution

Throughout this paper we have assumed that the parent distribution for various parameters is lognormal. This hypothesis is used for three purposes: (1) to combine the distributions of the four contributing factors to determine the distribution of f according to equations (4) and (5); (2) to determine the arithmetic means of the distributions of f and C_a according to equation (6); and (3) to estimate the fraction of households in which benchmark values of C_a are exceeded (Table 5). It cannot be proven that the parent distributions are lognormal. However, even if they are not strictly so, they may be sufficiently well approximated by the lognormal distribution that the results of our analyses constitute good estimates.

Considerable evidence supports the appropriateness of the lognormal distribution for representing f and C_w. As noted elsewhere, cumulative probability plots of the factors contributing to f suggest that the lognormal distribution may be appropriate. The objective Kolmogorov-Smirnov test demonstrates for the water-use rates and air-exchange rate that the hypothesis that the parent distribution is lognormal cannot be rejected at a very high level of significance. In addition, it can be demonstrated on theoretical grounds that the random multiplicative combination of independent factors yields a lognormal distribution in the limit of a large number of factors, regardless of the nature of the distribution of each factor. And finally, a previous study of radon in water found that the distributions by state were better fitted by a lognormal than by a normal distribution (Pr83).

Adequacy and Representativeness of the Data

Of the four factors needed to determine f, only the distribution of house volumes was obtained from a large random sample that may be considered representative of the United States housing stock. Of the remaining factors, we believe it is most important to improve information on in-house water-use rates. Even though the current data show only moderate dispersion, we are not assured that the limited sample of 90 residences is

at all representative of the entire country. The second priority for increasing the amount of data should be directed at the air-exchange rate. We are reasonably assured that no other large data sets currently exist; however, because of the recent development of infiltration models (*e.g.*, Gr82) and an integrating ventilation monitor (Di82), a systematic study of air-exchange rates in U.S. residences is now possible. The remaining factor, the use-weighted transfer coefficient, does not need much further consideration. Because any residence uses water in a mix of applications, some with high transfer efficiencies and others with low values, the use-weighted coefficient for any house will necessarily be intermediate between the minimum and maximum use-specific values of 0.3 and 1.0. The very narrow range for e observed, even for the limited data set considered here, combined with the good agreement among investigators on use-specific values suggests that further refinements in determining the distribution of e are unlikely to substantially affect the estimates of f.

Note that we have effectively taken the perspective that this fundamental approach -- based on data on four contributing factors and on a simple conservation-of-mass model -- provides sounder information on f than direct experimental measurement of airborne and waterborne radon concentrations. The limited data of the latter kind have an average and range consistent with those resulting from the analytical approach taken here. Improvements in the statistical representation of direct experimental determinations of f by measuring C_a and C_w are subject to considerable difficulties in terms of the measurements themselves, and formidable problems in choosing a sample where the water contribution is dominant, yet which is also representative of the housing stock.

Correlation Among Factors

The derivation of equation (1) assumed that the air-exchange rate, λ, and the incremental airborne radon concentration due to water use, C_a, were not correlated. No data exist to test the validity of this assumption. However, it is reasonable to expect, given the high proportion of water use in bathrooms and the common presence there of an exhaust fan or an open window, that these variables are correlated to a degree. Experiments would be needed to determine the importance of this possible correlation.

In determining variance in f, we further assumed that the several contributing factors were independent (see Equation 5). This assumption, while a reasonable approximation, is not strictly valid. For example, one study showed that water-use rates are positively correlated with house value (Li67); hence they probably are positively correlated with house volume. Data in another study show a negative correlation between house volume and air-exchange rate (Gr83).

The complete form of equation (5), accounting for correlation among factors, would include six additional terms, one for each pair of factors. The sign of each term can be plus or minus, depending on whether the correlation is positive or negative and whether one factor multiplies or divides the other in determining f. The only available data for estimating the magnitude of these terms are the measurements of house volume and air-exchange rate in 312 houses (Gr83). We computed the variance of the sum of the logarithms of these two factors, first assuming them to be independent, and then including the effects of cross-correlation. The cross-correlation term reduced the standard deviation of the sum by 11%. Since in determining the variance of f both negative and positive contributions from the cross-correlation terms are to be expected, the error in assuming the factors are uncorrelated is probably of order 10%. (Note that equation (4) for the geometric mean does not depend on the factors being independent.)

Limitations of the Model

In assessing population exposures using the long-term-average single-cell model, two additional elements must be considered. First, although the model assumes a uniform indoor concentration, in cases where water is a significant radon source for the entire house the concentration varies spatially, particularly during times when water use is high. As a result, in some cases the single-cell model may not effectively describe the spatially-averaged indoor concentration. For example, a bathroom exhaust fan may effectively prevent much of the radon released from a shower from reaching other rooms of the residence, even though it has a small impact on the air-exchange rate of the entire house. On the other hand, because of spatial association between occupants and the site of water use, the exposure to radon liberated by water is somewhat greater than the household average concentration. However, the effect of spatial variations on exposure is diminished by the mixing that occurs during the time over which the radon progeny concentrations increase to secular equilibrium. Prichard, in considering this effect, concluded that the *average* indoor concentration arising from water use resulted in considerably greater exposure than did locally elevated concentrations associated with episodic emission (Pr81).

The second factor to consider is the temporal association between occupancy and water use: most of the water use in a household occurs when people are present. As a result, the average exposure to radon progeny arising from the use of potable water is greater than the estimate obtained by multiplying the average indoor concentration from this source by the percent of time a person occupies the dwelling. The upper bound on

exposure (considering this factor alone) assumes the same average concentration and 100% occupancy.

SUMMARY AND CONCLUSIONS

Surface supplies of potable water do not contribute significantly to indoor radon concentrations. Groundwater supplies, which serve about 50% of the population, can in some circumstances constitute the predominant source. Public supplies derived from groundwater and serving 1000 or more persons have been extensively investigated; these are estimated to contribute an average of 2% to the mean indoor radon concentration in the affected houses. Private groundwater supplies appear to constitute a somewhat greater source; however available data are fragmentary, and further sampling studies are warranted.

The concentration of radon in indoor air in U.S. housing for a specified concentration of radon in water has been determined using a long-term-average, single-cell model and available data on house volumes, air-exchange rates, water-use rates and water-to-air transfer coefficients. The ratio of airborne to waterborne concentration so determined is represented by a lognormal distribution with a geometric mean and geometric standard deviation of 0.65×10^{-4} and 2.88, respectively. The statistical uncertainty in the geometric mean arising from limited sample size is 6%. This distribution is consistent with the results of direct measurements previously made in thirteen houses.

The limitations in the present analysis have been discussed. None are expected to have a major effect on the results. Refinements may be justified in further testing the validity of the lognormal distribution in representing f and C_a, in considering the effect of spatial and temporal variation in concentrations on estimating exposure, and in examining the effects of seasonal variations in air-exchange rate.

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Table 1. In-house water-use rates by function. For each study the first line gives summary results; subsequent lines, where present, give average results for specific households.

	Water Use by Function (10^{-3} m^3 $person^{-1}$ day^{-1})					
Reference[a]	Dish-washing	Shower/ Bath	Toilet	Laundry	Other	Total
Co74 (8 houses)	68.0	23.8	65.0	39.7		197
1		14.0	34.4	69.0		145
2		36.0	104	34.8		384
3		10.8	41.0			157
4		16.6	72.5			220
5		40.2	113			263
6		28.4	44.8	37.8		190
7		24.9	42.1	31.4		143
8		20.8	69.8	26.5		179
Li74 (35 rural)	11.6	41.9	68.2	51.9		174
Li74 (10 urban)	13.9	37.9	68.2	41.4		161
La74 (5 houses)	13.6	32.1	74.8	28.0	7.9	156
A	12.1	58.2	112	54	11.3	246
B	34.4	20.0	138	7.9	12.2	212
C	12.9	22.3	42.3	16.3	5.7	99
D	7.9	18.9	51.8	29.9	3.8	112
E	7.9	37.8	49.9	17.0	10.2	123
Be75 (5 houses)	14.0	32.9	55.6	43.8	21.9	168
1	4.6	13.1	68.7	55.4	38.5	180
2	1.3	31.8	38.8	26.8	17.8	117
3	7.3	61.7	89.9	20.9	49.7	230
4		29.6	108.4	59.8	93.0	291
5	3.7	40.9	37.8	34.3	19.1	136
Mi76	27	80	121	27	10	265
Re65[b]	14	76	91	32	10	223
EPA78[c] (11 houses)	19	38	35	40	20	152
A	18.9	51.0	34.4	47.6	62.3	214
B	10.2	30.2	32.9	8.3	15.1	96
C	13.2	27.6	21.2	49.5	35.5	147
D	18.9	50.3	31.8	28.7	25.7	155
E	10.6	32.8	31.0	52.9	29.9	157
F	18.9	21.5	23.4	48.0	15.5	127
G	14.0	32.9	24.6	15.9	25.4	113
H	11.0	22.3	34.8	42.3	77.8	188
I	23.0	26.8	29.9	61.2	16.6	158
J	20.8	46.1	52.2	35.9	15.9	170
K	26.8	44.2	41.2	49.1	53.7	215
Pa79, Ho84a	30 - 38	76 - 152	15 - 23	76 - 114		197 - 327

a Study sites: Co74 - Connecticut (4), Rhode Island - (2), California - (2); Li74 - Wisconsin; La74 - probably Connecticut; Be75 - Boulder, Colorado; Mi76 - estimated; Re65 - estimated; EPA78 - Wisconsin; Pa79 - estimated.

b Cited in USWRC78; apparently the basis for the analysis in Ge80 and Pr81.

c The basis for analysis in He82.

Table 2. Transfer coefficient for the release of radon from water to air, by use.

	Fraction of Radon Liberated			
Type of Use	Pa79	Ge80	He82	Mean
Dishwasher	0.98	0.9	0.98	0.95
Shower	0.71	0.63	0.65	0.66
Bath	0.5	0.47	0.3[a]	0.42
Toilet	0.29	0.3	0.3[a]	0.3
Laundry	0.95	0.9	0.9[a]	0.92
Drinking and Cleaning	0.28	0.45	0.1 - 0.5[a]	0.34

[a] Estimated in the original reference.

Table 3. Summary of the distributions for parameters used in the determination of the air-to-water ratio.

Parameter	N	GM	GSD	% Var.[a]	GSE[b]	Units
Water-Use rate (W)	90	7.9×10^{-3}	1.57	18	1.05	m^3 (person-hr)$^{-1}$
House Volume (V)	6051	98.7	1.90	37	1.01	m^3 person^{-1}
Air-Exchange Rate (λ)	578	0.68	2.01	44	1.03	hr^{-1}
Transfer Coef. (e)	21	0.55	1.12	1	1.03	
Air-to-Water Ratio (f)		0.65×10^{-4}	2.88		1.063	

a Percent of variance in $\ln(f)$ that can be attributed to given parameter.

b Geometric standard error: $GSE = \exp(\ln(GSD)/N^{1/2})$.
For f the GSE was determined as:

$$GSE_f = \exp\left[\ln^2(GSE_W) + \ln^2(GSE_V) + \ln^2(GSE_\lambda) + \ln^2(GSE_e))^{1/2}\right]$$

Table 4. Radon concentrations in public ground water supplies in the United States.

State	Population Served (Thousands)	No. Samples	^{222}Rn Concentration GM (10^3 Bq m^{-3})	GSD	Reference
Alabama	1200	104	3.29	3.10	EPA79c, EPA82b
Arizona	1490	64	10.9	2.40	EPA83
Arkansas	880	43	1.70	4.39	EPA79b, Pr83
Colorado	320	37	9.31	2.49	EPA82b
Delaware	254	36	3.56	1.88	EPA81a
Florida	6800	165	3.03	3.02	EPA81a
Georgia	1320	61	3.87	3.93	Ho84b
Idaho	592	85	8.30	2.41	EPA81b
Illinois	4050	158	4.95	2.09	EPA83
Indiana	1920	117	2.51	2.83	EPA82a, Pr83
Iowa	1600	58	4.37	3.23	EPA79b, Pr83
Kansas	903	7	2.66	3.11	HO84b
Kentucky	375	50	3.46	2.46	EPA81b
Louisiana	1850	22	3.44	2.54	Pr83
Maine	101	68	37.4	2.38	EPA79a
Massachusetts	1550	100	26.8	1.76	EPA82b
Minnesota	1910	124	6.72	2.17	EPA82b, Pr83
Mississippi	1800	53	2.10	2.61	EPA82b
Montana	184	33	14.0	2.17	EPA81b
Nebraska	961	21	6.59	3.68	Pr83
Nevada	329	26	13.3	2.28	EPA81a
New Hampshire	392	31	32.1	2.45	EPA80, EPA81b
New Jersey	3420	19	13.5	3.21	EPA80
New Mexico	798	89	7.50	2.32	EPA82a, Pr83
New York	3510	150	4.06	2.87	EPA79b
North Carolina	474	181	4.87	9.42	EPA79a, EPA82a
North Dakota	258	67	4.49	2.33	EPA81a
Ohio	2950	84	4.43	2.38	EPA81b
Oklahoma	662	56	4.82	1.97	EPA80, Pr83
Oregon	344	65	8.44	2.03	EPA81b
Pennsylvania	2180	89	14.2	3.22	EPA82a
Rhode Island	142	92	65.6	5.90	EPA83
South Carolina	541	185	5.03	6.28	EPA80
South Dakota	321	79	9.83	2.57	EPA81a
Tennessee	1450	50	1.24	5.64	EPA83
Texas	5030	278	4.85	2.70	Pr83
Utah	662	98	10.6	1.95	EPA82b
Vermont	113	11	23.1	1.48	EPA83
Virginia	707	101	8.37	3.69	EPA83
Wisconsin	1620	143	7.43	2.78	EPA82a
Wyoming	122	18	12.7	2.67	EPA83
Total	56,085[a]	3318	5.18[b]	3.53[b]	

a Includes 76% of population served by public ground water supplies.

b Population-weighted statistics.

Table 5. Estimated Contributions to U.S. Indoor Airborne Radon-222 Concentration for Three Types of Water Service.

	Fraction of U.S.	C_a (Bq m^{-3})			Fraction Exceeding:	
Type	Pop. Served	GM	GSD	AM	9.3 Bq m^{-3}	33 Bq m^{-3}
Surface	0.495	0.020	6.86	0.13	0.0007	0.0001
Public Groundwater	0.322	0.34	5.19	1.3	0.022	0.0027
Private Wells	0.183	2.3	8.59	24.	0.26	0.11
Aggregate	1.0			4.8	0.055	0.021

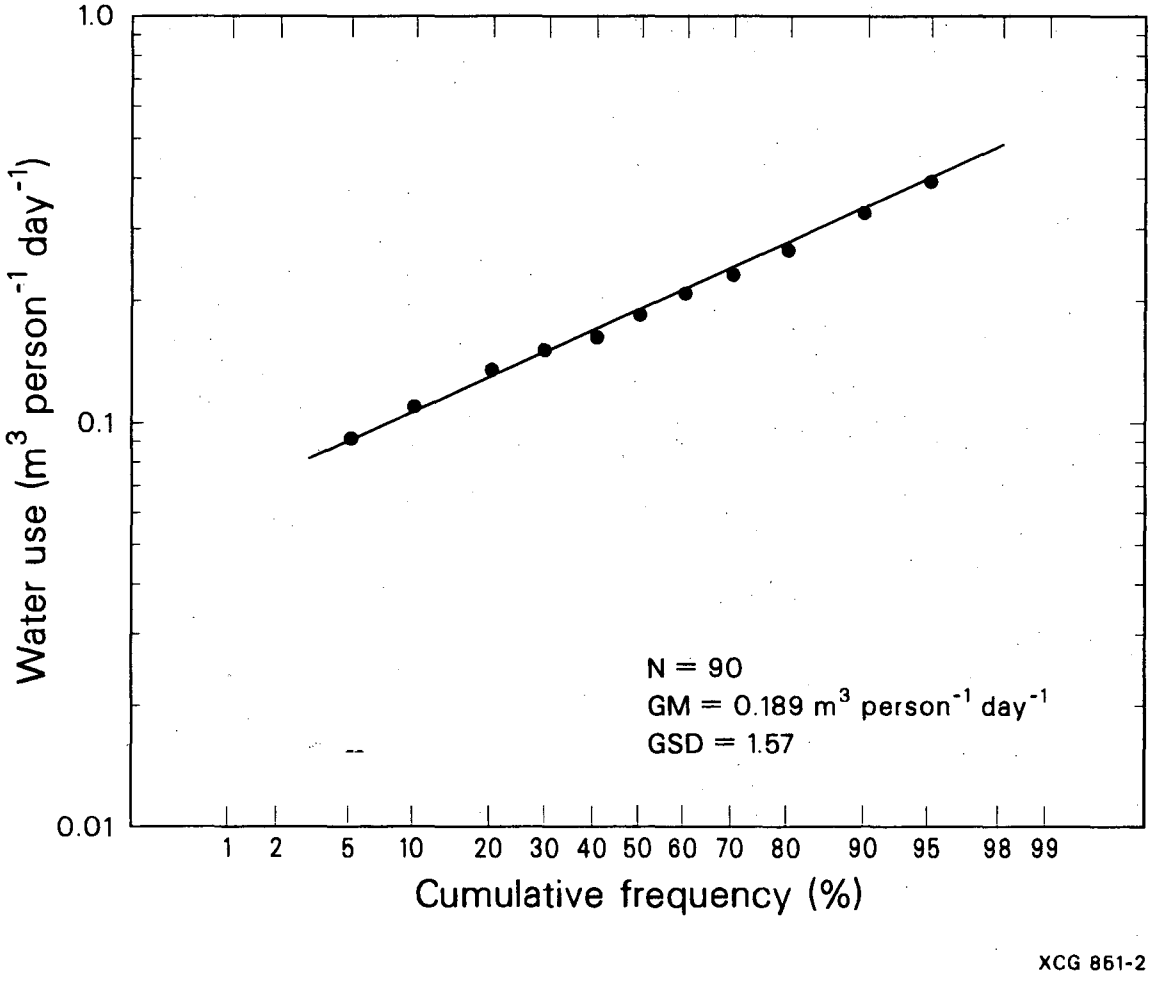

Figure 1. Cumulative frequency distribution of the average per-capita in-house water use rate, W, in 90 U.S. residences. The straight line represents the lognormal distribution that best fits the data, using minimum variance unbiased estimators of the geometric mean and geometric standard deviation.

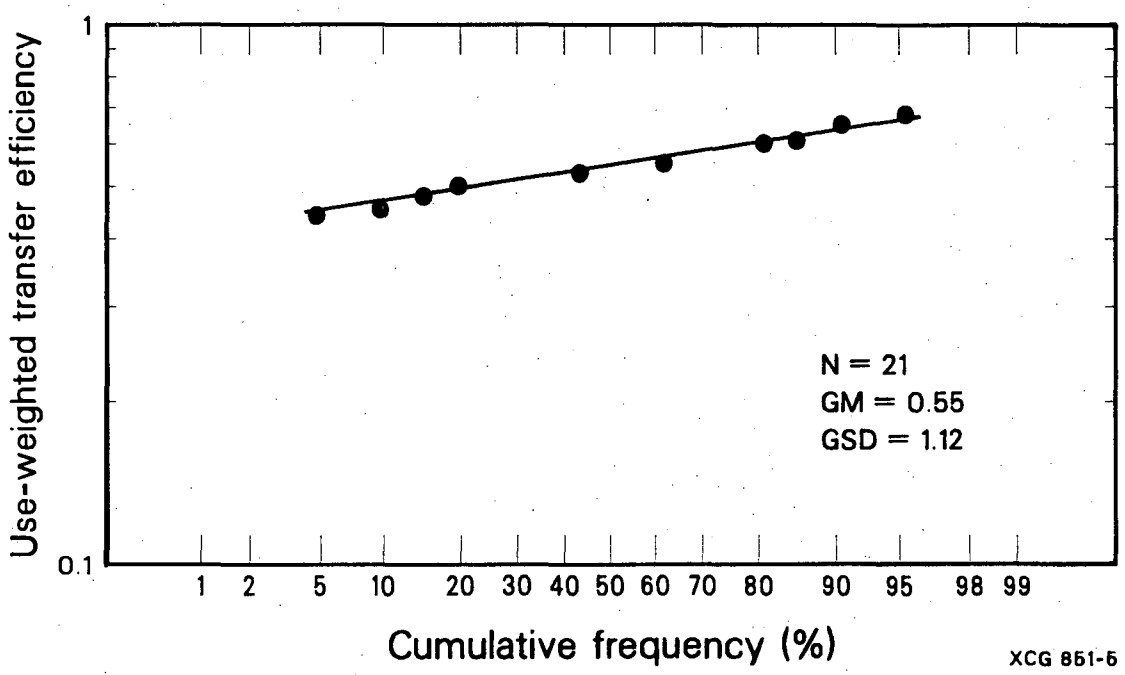

Figure 2. Cumulative frequency distribution of use-weighted transfer efficiency, e, for radon from water to air. Each of the 21 values was determined by summing the products of the use-specific transfer coefficients (Table 2, mean) and the fractional use rates for individual houses (Table 1).

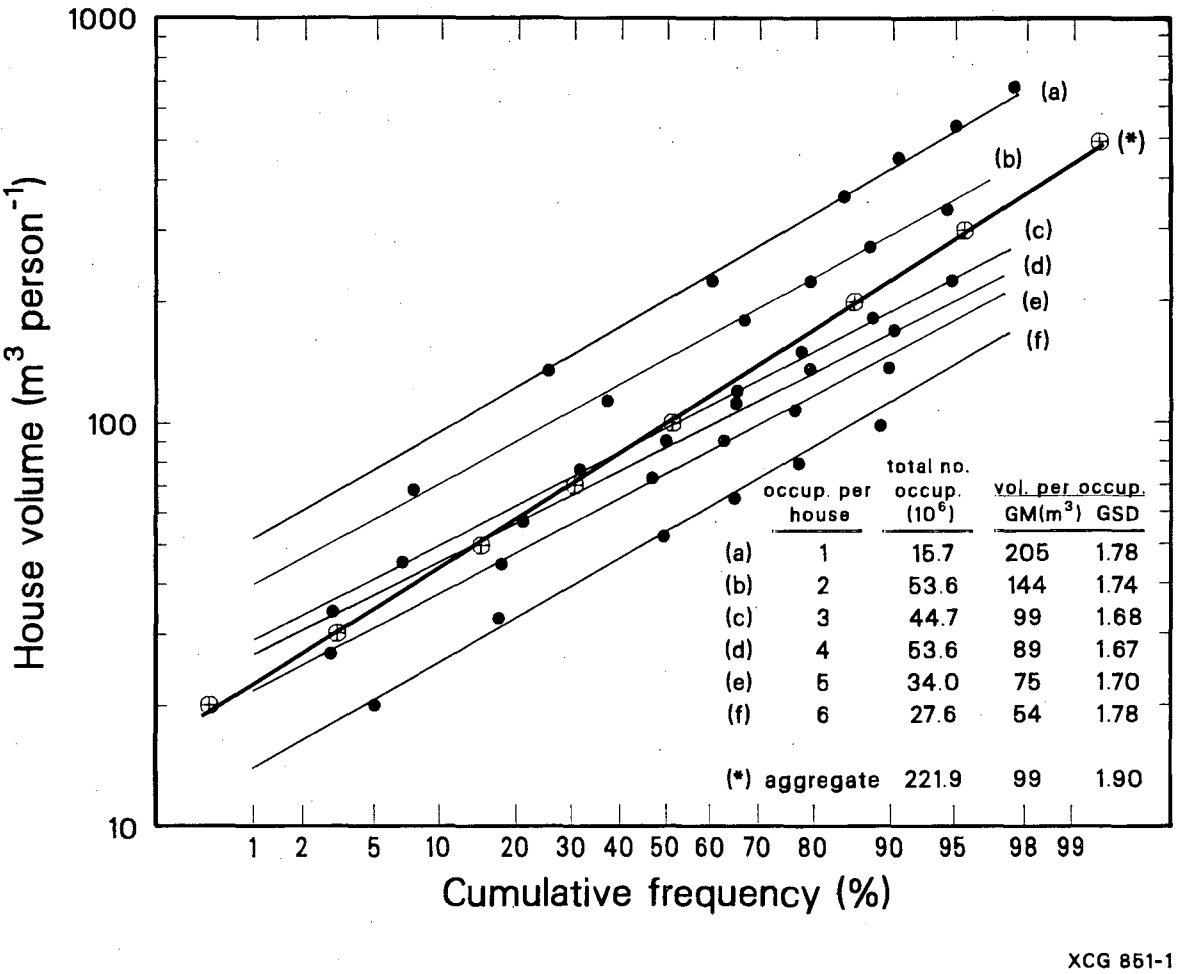

Figure 3. Cumulative frequency distribution of per capita household volume, V, for U.S. residences. Only heated volume is considered. Six curves, (a)-(f), give distributions for specified numbers of residents per household; the bold line gives the aggregate distribution, representative of the entire U.S. population.

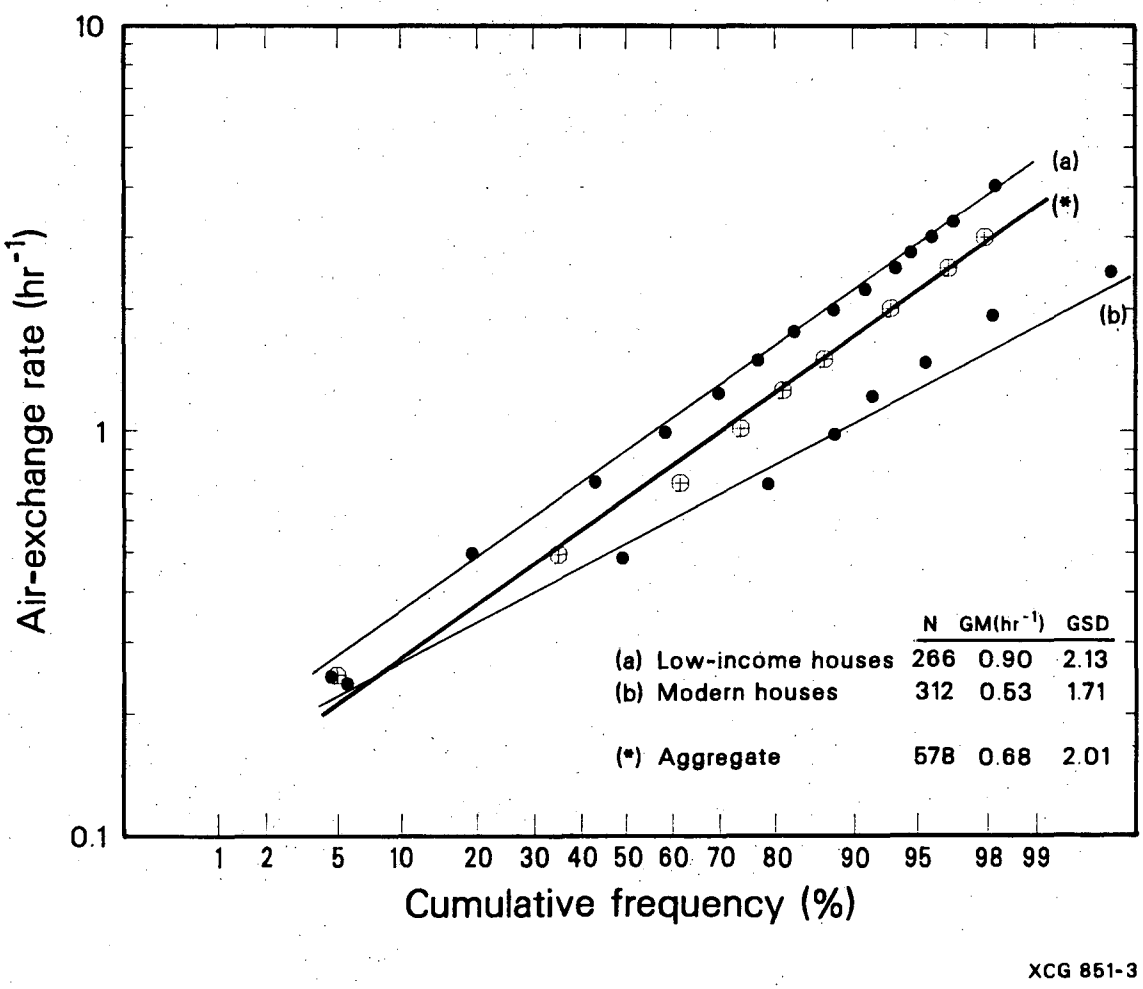

Figure 4. Cumulative frequency distribution of air-exchange rates, λ, in U.S. dwellings. The study of low-income houses measured air-exchange rates over intervals of a few hours (Gr81); the distribution was determined from 1048 individual measurements in 266 houses (and therefore has a higher variance than it would if the average measurement in each house had been used). The study of modern houses determined average infiltration rates for a heating season (Gr83). The studies were aggregated with weights proportional to the number of houses studied.

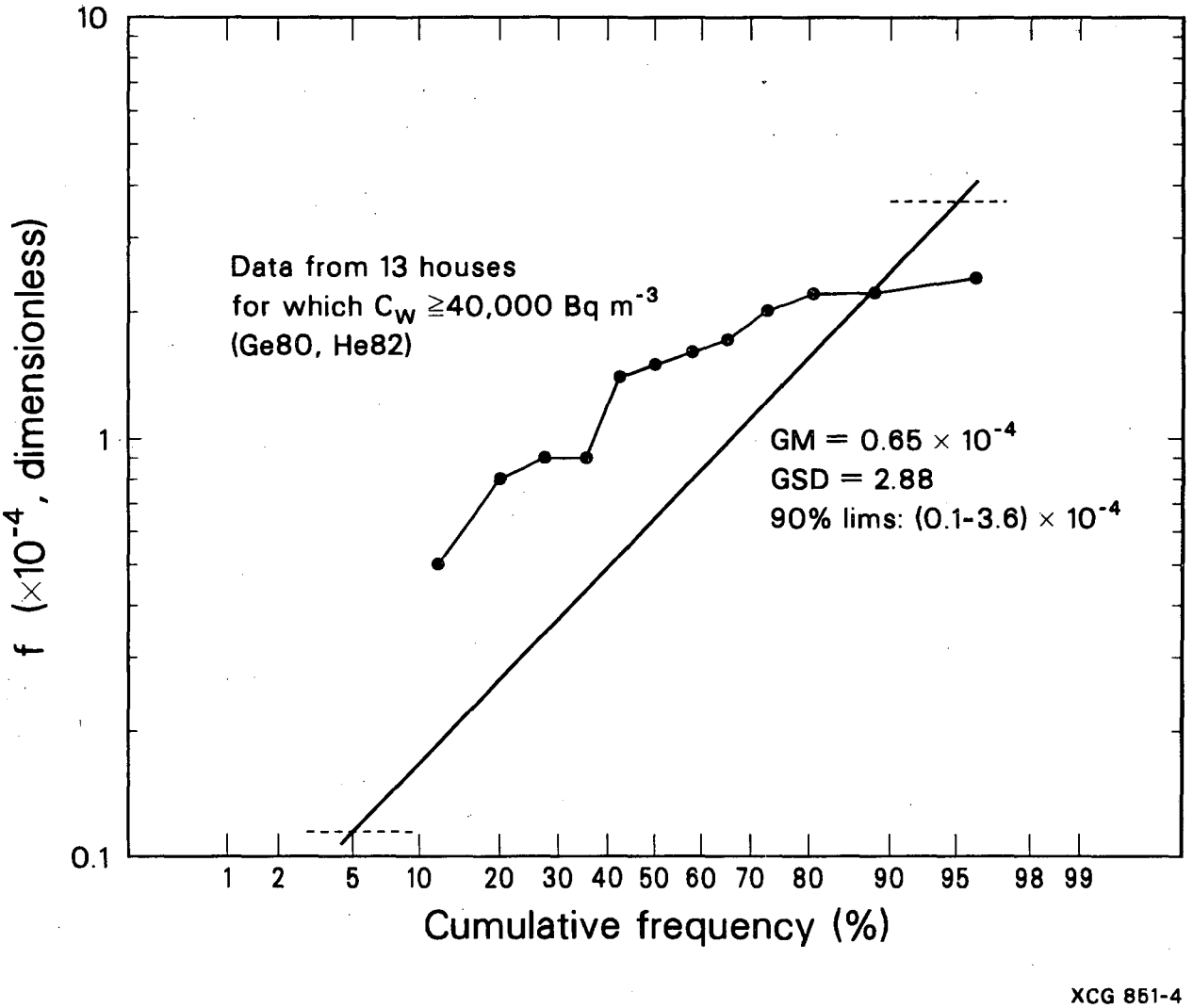

Figure 5. Cumulative frequency distribution of f, the airborne radon concentration in U.S. dwellings that can be ascribed to a specified waterborne concentration. The straight line represents the results of the analysis in this paper. The twelve points plotted represent results of measurements of f in twelve of thirteen houses for which the waterborne concentration exceeded 40,000 Bq m^{-3}. For the thirteenth house, a negative value was determined for f, which is not plotted in the figure. For three of the houses the reference reports C_a as less than a limit. For this figure we have plotted f for these houses at the corresponding limits, which are (0.8, 0.9 and 1.7) x 10^{-4}, respectively.

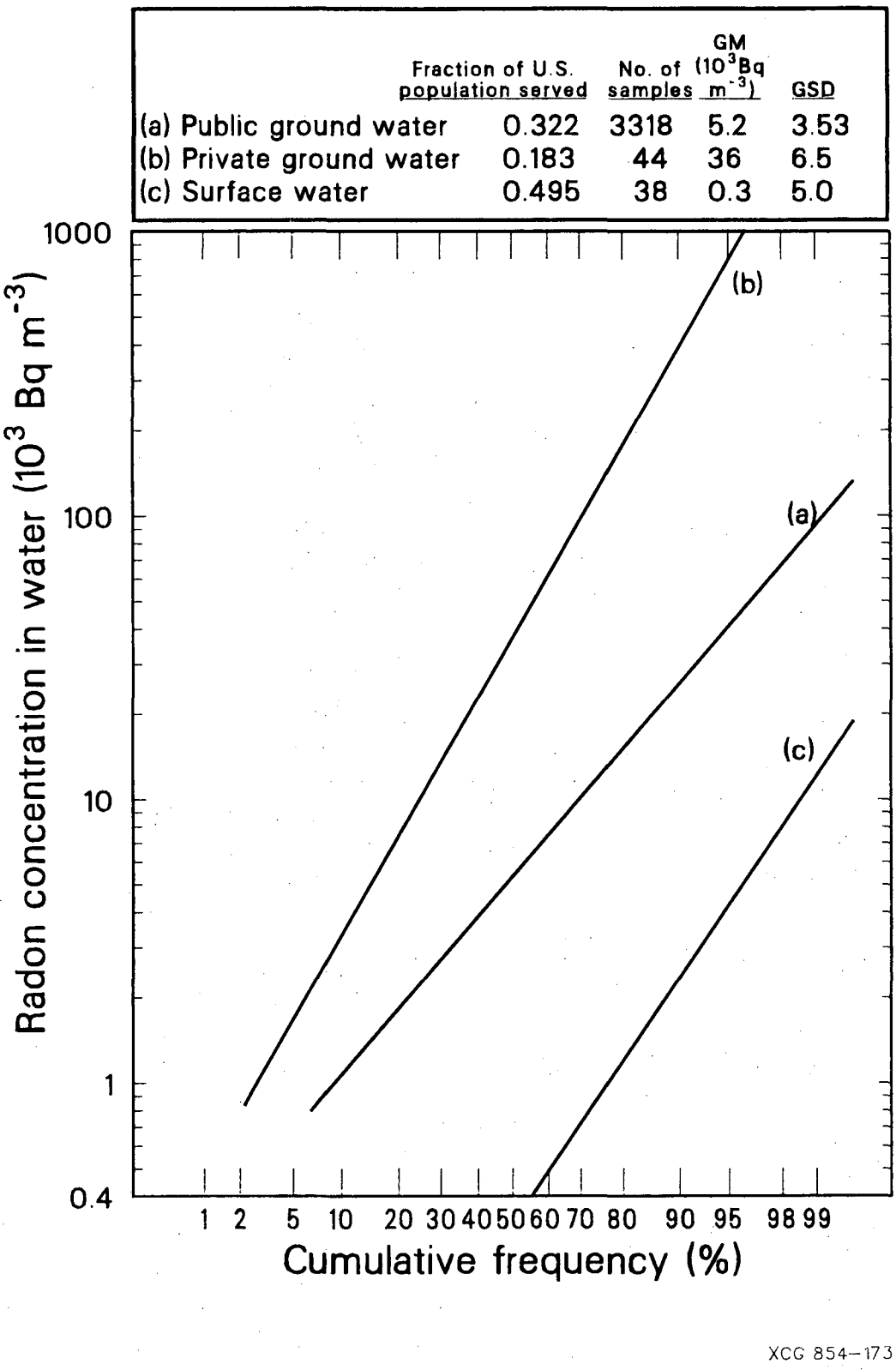

Figure 6. Cumulative frequency distributions of radon concentrations in potable water supplies in the United States. Distributions for surface water, public groundwater and private groundwater are plotted separately.

APPENDIX

Derivation of Equation (1)

By treating the interior of a residence as a single, well-mixed volume, the rate of change of the indoor radon concentration, C_i, may be described by the first-order differential equation

$$\frac{dC_i}{dt} = C_o\lambda^* + S - C_i\lambda^* + \frac{C_w W^* e}{V}, \tag{A1}$$

where C_o is the outdoor radon concentration,
S is the entry rate for all sources other than water (i.e., building material and soil),
λ^* is the instantaneous air-exchange rate,
C_w is the concentration of radon in water,
W^* is the instantaneous water use rate,
V is the volume of the residence, and
e is the use-weighted transfer efficiency of radon from water to air.

We have neglected radioactive decay as a removal term as it is small relative to the air-exchange rate.

If equation (A1) is integrated over a long period and divided by the length of the period, the left hand side tends to zero and we obtain the equation

$$\int_0^T C_i\lambda^*\, dt = \int_0^T (C_o\lambda^* + S)\, dt + \int_0^T \frac{C_w W^* e}{V}\, dt. \tag{A2}$$

Let $C_i = C + C_a^*$, where C is the indoor radon concentration in the absence of water use and C_a^* is the instantaneous concentration increase due to radon released from potable water. Then

$$\int_0^T C_a^*\, dt = \frac{1}{\lambda'} \int_0^T \frac{C_w W^* e}{V}\, dt, \tag{A3}$$

where

$$\lambda' = \frac{\int_0^T C_a^*\lambda^* dt}{\int_0^T C_a^* dt}. \tag{A4}$$

We define the time-average values, C_a and W, by the relations

$$C_a = \frac{1}{T} \int_0^T C_a^* dt, \tag{A5}$$

$$W = \frac{1}{T} \int_0^T W^* dt. \tag{A6}$$

Making the assumption that C_w, V, and e are constant, we obtain

$$C_a = \frac{C_w We}{\lambda' V}. \tag{A7}$$

In equation (1), we have replaced λ' by λ. This is equivalent to assuming that the air-exchange rate is not correlated with C_a. Alternatively, this assumes that the water-use rate and the air-exchange rate are not correlated.

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