1994 _ 20

VENTILATION RATE AND SOURCE EMANATION RATES BY MEASUREMENT OF INDUCED RADON TIME-DEPENDENT BEHAVIOR

Bobby E. Leonard
International Academy of Hi-Tech Services, Inc.
Annapolis, MD

ABSTRACT

The use of induced time-dependent Radon-222 behavior, to determine source rate magnitudes, ventilation rates (air change rates) and other parameters that affect Rn-222 and progeny levels and exposure to building occupants, is investigated. Under the conditions of purging the subject space and measuring the build-up (seepage) back into the space, theoretical, normalized equations show a unique, one-to-one correspondence of the Rn-222 and/or particulate progeny temporal levels to the air change rate in the space. The Bateman equations have been solved in closed form for Rn-222 and progeny in air and trapped on a filter under these conditions. A total of 28 measurements of Rn progeny time-dependent behavior for two test facilities (one with constant air change rate and one with a constant NIST-calibrated source) and four residential dwellings were made. The results were compared to theory and to air change rate measurements made by anemometer flow rates and by the conventional method (SF_6 decay). Over a factor of 2 range in air change rates in the NIST constant source case the agreement with the SF_6 method air change rate was within $\pm$ 10.6% S.D. and agreement with the NIST source magnitude of 37.0 $\pm$ 1 kBq was within $\pm$ 4.9% S.D. Agreement to within $\pm$ 17.7% S.D. was obtained on determination of air change rates for the residential dwellings. Knowing the airborne concentrations and air change rates, source emanation rate magnitudes are obtained.

INTRODUCTION

Radon mitigation in residential spaces can be successful in many instances by improving the ventilation rate or a combination of ventilation increase and producing a positive pressure differential between the occupied spaces and the Rn source, if ventilation rate is known. The EPA Report 625/5-87/019, "Radon Reduction Technique for Detached Houses"[1] explicitly states that "the most effective measure that a homeowner can take to reduce radon levels" is to increase the ventilation rate. The report further specifies measurement of house ventilation rate as a diagnostic test to aid in the selection and design of Rn reduction techniques.

Even so, a local survey of 47 Rn testing and mitigation firms in the State of Maryland (involving 90% of those licensed) found that only two firms had the capability, in-house, to measure ventilation rates. A common method of performing ventilation rate measurements is to thoroughly mix a tracer gas, such as sulfur hexafluoride or ethane, with the indoor air, and to measure its rate of decrease in concentration by escape from the space. From the survey of the Rn firms contacted, it appears that ventilation measurements, by this conventional method, being a non-nuclear type assessment, is not being offered by them as a diagnostic option since it requires extensive tracer gas analysis equipment (cost $10-20,000) and also perhaps because it is out of their realm of nuclear expertise. If it were possible for the Rn tester and/or mitigation contractors to adequately measure Rn level and air change rates with a single Rn detection instrument, perhaps more measurements would be made and, in such cases, a better assessment of the mitigation options would be possible. This work demonstrates a definite feasibility.

BACKGROUND

The relation $C \sim S / \lambda$, is a simplified, stationary, equilibrium, result from the Bateman (1908) equations (where C = radon concentration in Bq m^{-3}, S = radon emanation rate magnitude in Bq h^{-1} m^{-3} and λ = total removal

rate in h[-1]). Evans (1955) has examined the Bateman equations extensively for series of radioisotopes and Evans (1969) and Bigu (1985) have applied them to time dependent Rn-222, and Rn-220 and their progeny behavior in underground mines. Nazaroff et al (1988) and Nero et al (1983) have applied the stationary solutions to measuring Rn source magnitudes in residential buildings. Many others have used stationary relations in the study of infiltration. Of more direct relevance, Hess et al (1982) used time-dependent Rn progeny behavior to determine residential air change rates. In this case, the injection of a "spike" of Rn associated with the brief use of showers in the houses, represented the injection of a "tracer" gas and by assuming a correlation of its time-dependent decrease, through the Bateman equations, with building air change rate, they obtained approximate ventilation rates. Terilli and Harley (1987) obtained indoor ventilation rates from natural Rn-222 burst events using time series plots of the decrease in the Rn-222 concentration after these burst events when they occurred spontaneously.

The following sections will present the method whereby a Rn testing technician or a research scientist can routinely use the theory developed here to measure air exchange rates in confined spaces, to an accuracy equal to or better than present conventional methods, using currently available nuclear radiation detection instruments. This method involves the measurement of the buildup of the radon progeny following purging of the space being measured. The theory for this method is presented elsewhere (Leonard, 1993). Procedural requirements dictated by the theory will be discussed here. Then a series of experiments will be described that apply and evaluate the method. This will include the equipment and facilities used. Finally, the results and evaluation will be given. In this text, air change rate will be denoted by ACH in units of h[-1]. The method will be called the Rn method and on occasion in the text we will refer to Rn and its daughters as just Rn when generalities are involved. The statistical treatment herein is that given by Evans.[3]

APPLICATION OF THEORY

To illustrate the method, we refer to Fig. 1. In measuring the buildup of the radon progeny activity on a filter, $A_f(t)$, it is necessary to normalize $A_f(t)$ such that $A_f(t)/A_f(\infty)$ is independent of the Rn and progeny source rate magnitudes. The asymptotic, equilibrium $A_f(\infty)$ must be experimentally determined. Therefore, the stationary Rn progeny level must be measured (in Region A) just prior to the purging (in Region B). It is desirable that sufficient counts be obtained in Region A to yield as good a statistical accuracy as possible. It is also desirable to limit the count time to minimize the chance of change in the Rn source level. At time point 1, purging should begin utilizing blowers and portable, flexible, plastic, lightweight ducts. The blower air removal rate should be such that the air volume in the space is exchanged about once every 5 minutes. The purging should continue for about 30 minutes, which would provide a total of 6 air turnovers resulting in a reduction of Rn and progeny levels in the space by a factor of 64, to less than 2% of the equilibrium level found in Region A.

Immediately before purging is stopped, the filter in the air sampler must be changed and at the moment that purging is terminated, the air pump, the detector and a clock (timer) must be started. All conditions in the space should be restored as to exactly what they were in Region A, such that the Rn and progeny levels will, in time, return to the equilibrium levels. This is at time point 2 which marks the beginning of Region C, to track the buildup of the Rn and progeny emanating back into the space. The rate at which the activity builds back up to its original level found in Region A is directly related to the ACH in the space. There is a one-to-one correspondence, between the normalized activity, $A_f(t)/A_f(\infty)$, at any time during the buildup as it approaches unity, to the air change rate in the space. The vertical lines in Fig. 1 show a read time interval of every 15 minutes for a period to 2.5 hours after the completion of purge at time point 2. The total elapsed time from the beginning of the initial stationary count in Region A, to the end of Region C at time point 3, is here given to be 3 hours, 10 minutes. This provides, in Region C, 10 experimental time data points to compare to theory to obtain a statistically significant measure of air change rate.

The normalized time-dependent buildup is obtained by dividing the $A_f(\infty)$ acquired in Region A into the individual activity values acquired in Region C. To correlate experimental $A_f(t)/A_f(\infty)$ (count rate) and $I_f(t)/A_f(\infty)$ (integral) values with theoretical ACH, two sets of two dimensional tables were computed respectively providing

count rate and integral values for times after purge from 0 to 5.0 hours at 0.25 hour intervals and for ACH values from 0 to 1.5 h⁻¹ at 0.01 h⁻¹ intervals. Table 1 provides excerpts from the integral table; each column represents the variation of the dependent variable [$(I_f (t) / A_f (\infty))$] with time for fixed ACH values, and each row provides for the variation of the dependent variable with ACH for fixed time after purge. By interpolation, for any accurate, measured values of A_f or I_p and $A_f(\infty)$, ACH as predicted by theory may be determined from Table 1 to ± 0.002 h⁻¹. It is noteworthy that since activity on the filter of the detector and counts rate on the detector counter defer only by the counting efficiency factor of the instrument, Count Rate (t) / Count Rate (∞) = $A_f(t)/A_f(\infty)$, and Integral Count (t) / Count Rate (∞) = $I_f(t) / A_f(\infty)$, such that when normalized values are used; normalized Count Rate (t) and Integral Count (t) are synonymous with normalized $A_f(t)$ and $I_f(t)$ in this text.

MEASUREMENTS AND RESULTS

Test Chamber--Natural Source, Mechanical ACH

Eight separate ACH values have been measured in a test chamber with a natural source emanation and no natural ventilation (less than 0.005 h⁻¹ ambient natural air change rate as determined by SF_6 tracer measurement). The chamber consists of 30 cm thick concrete walls, ceiling and floor with the only opening being a 60 cm by 100 cm hatchway. It is completely underground with 1 meter of earth cover over the ceiling slab. From prior measurements it was known to have a high natural Rn source level. In this chamber, a constant air exchange rate was induced mechanically with a box-type fan and plastic ducts. The eight different air change rates were established in the chamber and monitored by an anemometer at specific grid points across the face of the duct. Data was acquired for a minimum of 2.5 hours after purge. Both integral counts and count rate data were analyzed. For Run No. F-1, the equilibrium count rate $A_f(\infty)$, obtained was 1456 counts per hour. For nine 0.25 h interval values, (0.5 to 2.5 h), a value for ACH was computed from interpolation of the tables and for each data point a standard deviation (S.D.) from the mean value of all nine values was obtained and the S.D. was computed for the set and given in Table 2 Part A. Table 2 Part A provides results of the entire eight ACH measurements made with the mechanically induced ventilation in this chamber.

Test Room -- Constant Radon Source, Natural Building Ventilation Rates

Measurements were made by the same procedure described above, in a room on the upper floor of a building (where there was confirmed natural Rn level of less than 1.8 Bq m⁻³) using an NIST-calibrated 37k Bq (1.0 μ Ci) Ra-226 source. The natural ventilation of the space was present but with no forced (mechanical) ventilation. In this series of 3 measurements, the ACH was measured, simultaneous with the Rn and progeny buildup time (Region C), using both the Rn method and the conventional SF_6 tracer gas method with an Ion Track Model 61 SF_6 detector. The Rn progeny detector used in this series was a Victoreen Alpha CAM filter airborne radioactivity detector which has a much higher detection sensitivity. Values of ACH were obtained for each time data point after completion of purge. From these data, mean values for ACH by both methods were computed and a S.D. of each set of data was obtained (CAM S.D. and SF_6 S.D.). Table 2 Part B provides these data for the three measurements. Knowing the room volume (V) as 34.85 m³, we obtained values for the source rate magnitudes, through the relation S/V = C∞/λ, of 38.9 ± 4.7, 38.1 ± 8.4 and 34.8 ± 4.3 k Bq for an average of 37.3 ± 6.2 k Bq (1.01 ± 0.16 μ Ci) as compared to the NIST-quoted value of 37.0 ± 0.7 k Bq (1.00 ± 0.02 μ Ci).

Residential Measurements

A total of 17 ACH measurements were made in four single-family, detached houses with basements located in Annapolis, Maryland. All measurements were made in the basements with all air circulating systems shut off. In all of these measurements, simultaneous measurements of ACH were made by the SF_6 method and this Rn method and the same technique of computing the mean value and S.D. was used as described for the Constant Source test room (section B above). Table 2 Parts C, D, E and F provide the results. For the four dwellings, a wide range of natural air change rates were observed; i.e., from 0.175 to 2.741 h⁻¹.

To illustrate the form of the acquired data, we provide Fig. 2 for Run No. S-2. Fig. 2 provides experimental count rate (2a) and integral (2b) data and the theoretical curves for Rn progeny buildup for the mean value of ACH

= 0.531 h⁻¹. Counts were obtained each 2 minutes for these curves. Fig. 3 provides all data obtained for determination of ACH during Run No. S-2, showing 1.) the experimental SF_6 decay curve and the theoretical assuming a pure exponential decreasing with the SF_6 mean value ACH = 0.536 h⁻¹; 2.) the experimental integral curve for the Rn method buildup and the theoretical curve for the Rn method mean value ACH = 0.531 h⁻¹; and 3.) the ACH values computed for each time data point for 0.5 to 2.5 h by the SF_6 and Rn methods. From these time data point values, shown by the markers, the mean values of ACH = 0.536 and 0.531 h⁻¹, respectively, were computed with the % S.D.'s as given in Table 2 Part E. This method was used to determine the ACH's and their % S.D. for all values in Table 2 Parts C, D, E and F.

RESULTS AND DISCUSSION

Various facets affecting the accuracy of the method, such as counting statistics, instrument sensitivities, variation in source magnitude, plateout and incomplete purging, are discussed in an earlier paper (Leonard 1993). The objective of the measurements in the underground chamber was to test the theory and method under constant, known ACH values but where the experiments were susceptible to large variation in natural source rate magnitudes. Table 2 A provides the data obtained in the F-1 through F-8 Runs for the natural source, mechanical ACH case. From Table 2 A, the net % S.D. is ± 26.8% for the variation of ACH for the 8 sets of 9 time data points from each run using the Integral Table 2 values of ACH vs Normalized Integral Count for the time data points. In comparing the measured ACH by the Rn method to the "True ACH," the data provides ± 14.3% S.D.

The purpose of the Constant Source Room measurements were to test the theory and method under conditions where the source rate magnitude remained constant and the ACH was allowed to vary from natural building effects. From previous measurements, it was known that this room was susceptible to the diurnal variation in ACH from temperature effects and also variation from wind effects. As a result, the three measurements provided a range in natural ACH of a factor of 2. As indicated earlier, ACH was measured by the SF_6 method, also. By computing the variances for each 9 sets of data for both the Rn and SF_6 methods, net % S.D.s of ± 16.5 and ± 20.9% were obtained, respectively. Table 2 B provides the results for Runs A-1 through A-3 by the SF_6 and Rn methods. In comparing the Rn method to SF_6 method values obtained, we obtain agreement to within ± 10.6% S.D. Since the source rate magnitude was known to be 37 k Bq (1.0 μ Ci), it was possible, from the equilibrium concentration (C∞) measured with the Alpha CAM during the three runs, to compute an experimental source rate magnitude for each run providing values of 38.9, 38.1 and 34.8 k Bq. By computing the variances with respect to the calibration value for the Ra-226 source, we obtain ± 4.9% S.D.

The residential measurements utilized both the SF_6 and the Rn method, also. Fig. 4a provides a bar graph with the individual runs noted and the values from the two methods adjacent to each other. Also, as Fig. 4b, we provide a graph of SF_6 method ACH vs Rn method ACH. The diagonal line is the 1:1 ratio line. The net % S.D. for each method from the variance of the time data points was ± 31.1% S.D. and ± 34.5% S.D. for the Radon and SF_6, respectively. If the data for Run No. W-4 is rejected, due to the occurrence of a strong weather front passing through during the measurements (47 km h⁻¹ winds), we obtain ± 21.6% S.D. and ± 23.6% S.D., respectively. In comparing the ACH values obtained, by computing the variances between the ACH values for the two methods, we obtain a ± 17.6% S.D.

SUMMARY

The feasibility of using temporal variation of Rn and progeny to measure air change (ventilation) rates in buildings has been examined. A theory was developed using the Bateman equations assuming constant Rn and progeny source rate magnitudes and constant air change rate in the space. The measurement sequence was 1.) determine the equilibrium Rn and progeny level in the space and if found to be equal to or above EPA action level, then 2.) purge the space of the Rn and progeny bearing air and 3.) monitor the return of the radioactivity back into the space. Rigorous solutions to the equations are provided with analytical and digital form results. In comparing

experimental measurements with theory, a one-to-one correspondence exists between the returning Rn and progeny level in 3.) above to space air change rate.

Measurements have been made in an underground test chamber with natural Rn and progeny entry source rates and no natural ventilation but imposing known forced ventilation rates. Agreement is found to within ± 14.3% S.D. Measurements have been made in an above-ground test room using a constant NIST-calibrated Ra-226 source and natural ventilation by both this method and the conventional SF_6 tracer gas method. Agreement is to within ± 4.9% S.D. to the NIST source rate magnitude value and agreement to within ± 10.6% S.D. between the SF_6 and Rn method values. Seventeen separate measurements were made in the basements of four separate single-family dwellings obtaining agreement between the SF_6 and Rn methods to within ± 17.6% S.D.

The method provides a means for scientists and Rn testing and mitigation personnel to determine air change rates directly with Rn and progeny monitoring devices. Complete tables of $A_f(t) / A_f(\infty)$ and $I_f(t) / A_f(\infty)$ as function of time and ACH are available as an internal report (International Academy, 1991). For the benefit of the tester/mitigator, this work indicates the feasibility of developing a field radiation detection instrument similar to the Victoreen Alpha CAM with all the theoretical equations and tables for the Rn method included, on-board, in a micro-computer. Such an instrument has been designed and is available by special order from Victoreen. A field technician with minimal training should be able to perform the Rn measurements of Region A and Region C, Fig. 1. With appropriate switches and buttons, this instrument would automatically perform all data collection and analysis and then output to the technician the Rn progeny concentration, air change rate, source emanation rate and the % S.D. accuracy of each. The primary duty of the technician would be to set up the equipment, properly purge the space and change the filter.

REFERENCES

U.S. Environmental Protection Agency. Radon reduction techniques for detached houses. (EPA/625/5-87/019): (1988).

Bateman, H. The solution of samplings of differential equations arising in the theory of radioactive transformation. In: Proceedings of Cambridge Philosophical Society, 15: 423: 1908.

Bigu, J. Theoretical models for determining ^{222}Rn and ^{220}Rn progeny levels in Canadian underground U mines - a comparison with experimental data. Health Physics Journal, 48: 4: 371: 1985.

Evans, R.D. The Atomic Nucleus. New York: McGraw Hill Book Company, Inc.: 1955.

Evans, R.D. Engineers guide to the elementary behavior of radon daughters. Health Physics Journal, 17: 229: 1969.

Hess, C.T. et al. Variations of airborne and waterborne Rn-222 in houses in Maine. Environmental International, 8: 59: 1982.

International Academy of Hi-Tech Services. Ventilation rates by direct measurement of temporal Rn behavior. Report IA 91-145: 1991.

Leonard, B.E. Ventilation rates by measurement of induced radon time-dependent behavior - theory, application and evaluation. Nuclear Tech., 104: 89: 1993.

Nazaroff, W.W., et al. Radon transport into a detached one-story house with a basement. Atmospheric Environment, 19: 1: 31: 1988.

Nero, N.V. et al. Radon concentrations and infiltration rates measured in conventional and energy-efficient houses. Health Physics Journal, 45: 8: 401: 1983.

Tercilli, T.B.: Harley, N.H. Indoor ventilation rates for ^{222}Rn. Health Physics Journal, 52: 6: 801: 1987.
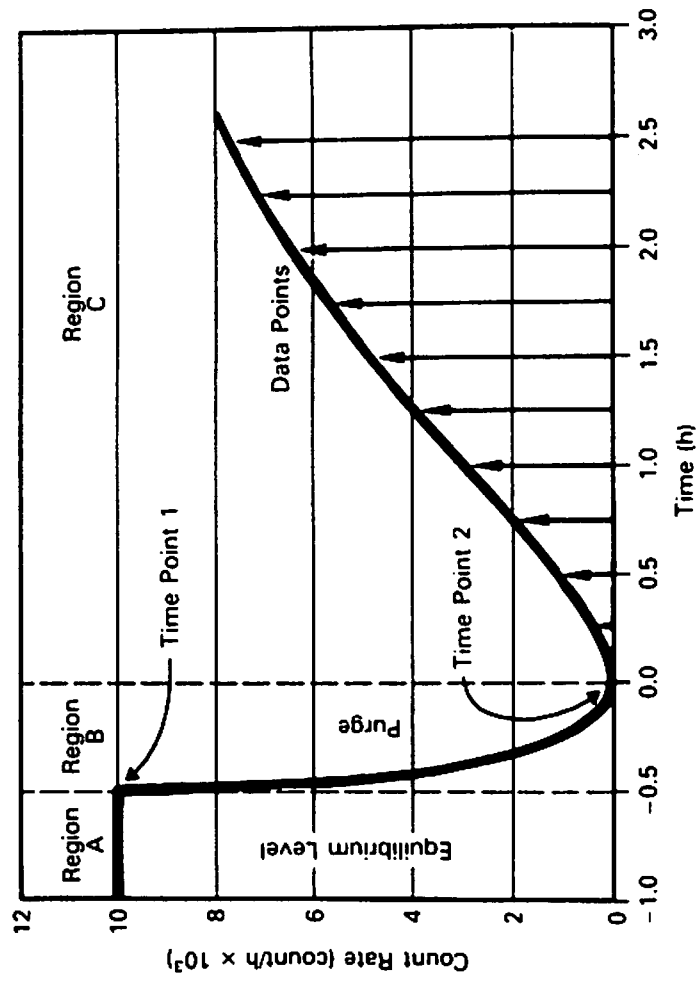
Fig. 1. Graphical illustration of measuring sequence, giving counts per hour as a function of time after purge, showing equilibrium region (region A) with hypothetical stationary value of 10,000 count/h, purge region (region B) of 30 min, and buildup region (region C) where count rate data are collected every 0.25 to 2.5 h.
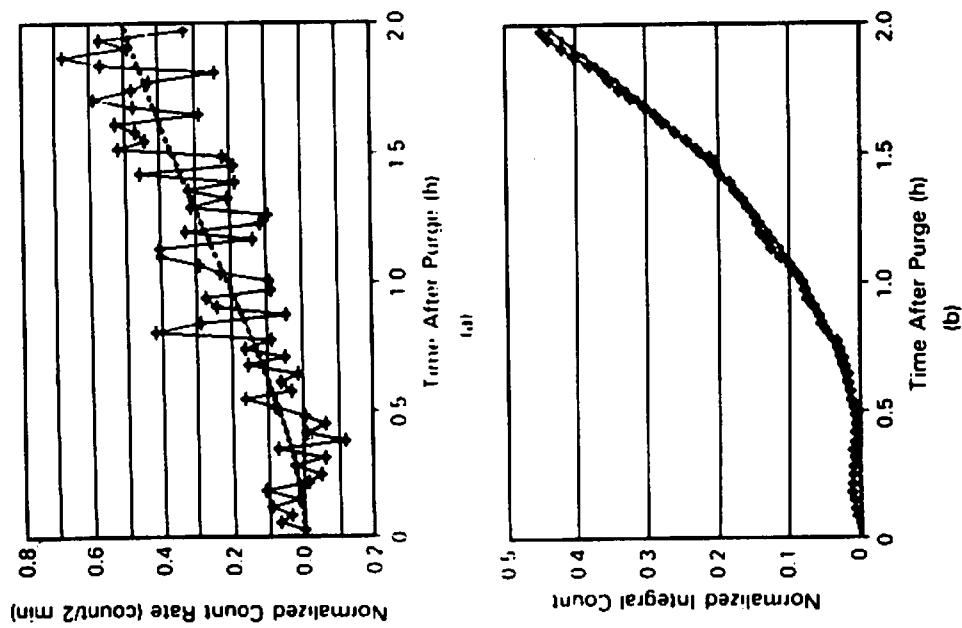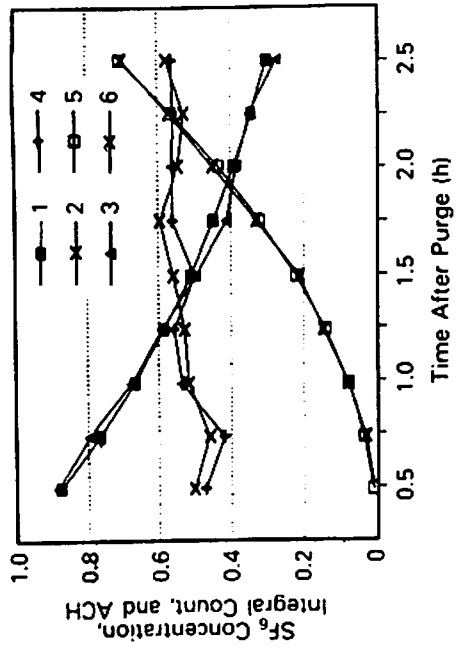
Fig. 3. Comparison of ACH time data points for the SF$_6$ tracer gas method (curve 2) and the radon method (curve 4) for run S-2 as a function of time after purge. Also, experimental (curve 3) and theoretical (curve 1) SF$_6$ concentration decay and experimental (curve 5) and theoretical integral count (curve 6).

Fig. 2. Experimental data (+) for run S-2 for (a) count rate and (b) integral counts and theoretical curves (*) for ACH = 0.531 h^{-1}.
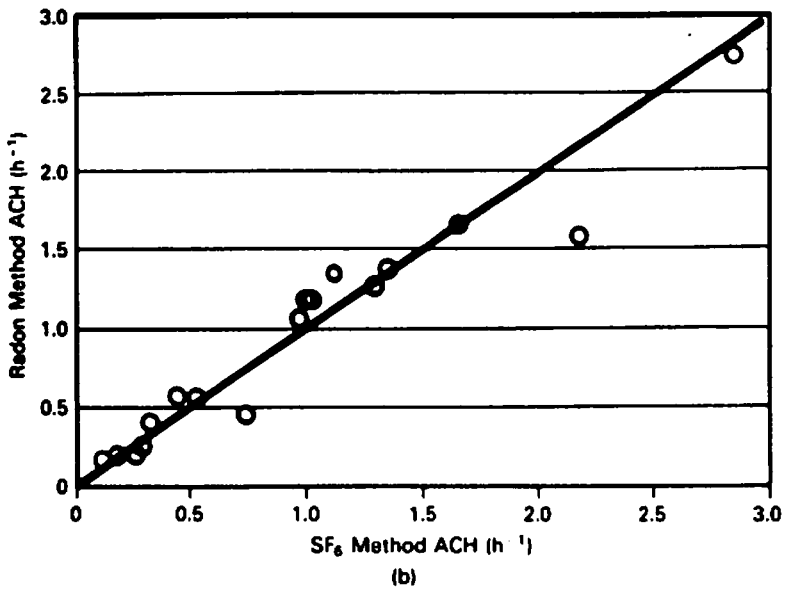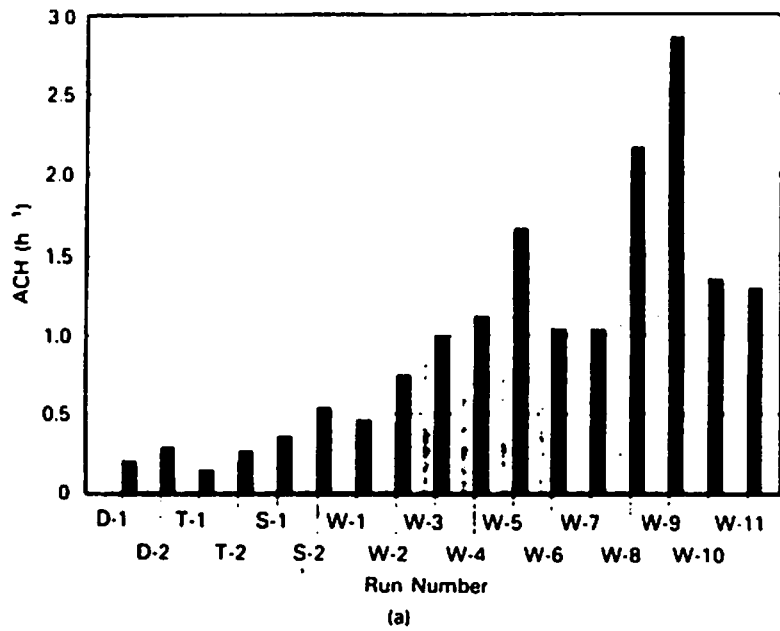
Fig. 4. Residential measured ACH values: (a) bar graph of ACH values by the SF₆ method (left) and the radon method (right) and (b) same data on an X-Y plot.

Table 1

Excerpt from Table of Theoretical Integral Count as a Function of Time After Purge and ACH, Used in Determining ACH for Discrete Measured Time Values and Values of $I_f(t) / A_f(\infty)$ by the Radon Method

Time (h)	ACH (h⁻¹)			
	0.01 $I_f/A_f(\infty)$	0.02 $I_f/A_f(\infty)$	1.49 $I_f/A_f(\infty)$	1.5 $I_f/A_f(\infty)$
0	0	0	0	0
0.25	4.47E−05ᵃ	8.59E−05	0.004729	0.004755
0.5	0.000307	0.000591	0.029787	0.029936
0.75	0.000936	0.001799	0.083649	0.084028
1	0.002041	0.003918	0.168745	0.169438
1.25	0.003694	0.007088	0.283883	0.284941
1.5	0.005945	0.0114	0.425941	0.427387
1.75	0.008823	0.016907	0.591025	0.592852
2	0.012344	0.023639	0.775159	0.777345
2.25	0.016519	0.031612	0.974666	0.977177
2.5	0.02135	0.04083	1.186342	1.189138
2.75	0.02684	0.051294	1.4075	1.410542
3	0.032987	0.062999	1.635954	1.639202
3.25	0.039792	0.07594	1.869961	1.873381
3.5	0.047251	0.090111	2.108159	2.111718
3.75	0.055362	0.105505	2.349492	2.353164
4	0.064125	0.122116	2.593156	2.596917
4.25	0.073536	0.139936	2.838542	2.842375
4.5	0.083594	0.158959	3.085195	3.089083
5	0.105644	0.200589	3.58103	3.584994

ᵃRead as 4.47 × 10⁻⁵.

Table 2

Experimental Data for Measurements Using Time-Dependent Radon Progeny Data to Determine ACH*

Constant Forced ACHR, Natural Source Underground Chamber

Mechanical ventilation, chamber volume = 43.65 m³
Chamber underground surface area = 83.4 m²

1	Sniffer Data			Anemometer Data	
	2	3	4	5	6
F-1	1456	0.205	54.1	0.157	2
F-2	3840	0.345	13.4	0.291	2
F-3	9600	0.035	6.3	0.033	2
F-4	1452	0.354	8.1	0.357	2
F-5	13500	0.067	23.8	0.075	2
F-6	6600	0.161	42.9	0.177	2
F-7	61050	0.00018	12.4	0.000	2
F-8	63010	0.104	3.5	0.101	2

Net percent standard deviation = 26.8

Constant Source Test Room

NIST-calibrated 226Ra source = 37.0 kBq
Underground surface area = 0.0 m²
Room volume = 34.85 m³

1	CAM Data			SF₆ Data	
	2	3	4	5	6
A-1	6287	0.119	12.1	0.118	25.2
A-2	2923	0.251	22.8	0.244	24.6
A-3	3045	0.221	12.5	0.205	8.3

Net percent standard deviation = 16.5

Residential Measurements

Natural Source, Natural Ventilation

1	CAM Data			SF₆ Data	
	2	3	4	5	6
House D — Volume = 285.55 m³, Underground surface area = 205.2 m²					
D-1	19872	0.213	22.1	0.202	12.2
D-2	4022	0.252	3.9	0.289	9.2
House T — Volume = 110.14 m³, Underground surface area = 101.1 m²					
T-1	240	0.175	18.7	0.142	14.7
T-2	3855	0.197	8.6	0.262	39.8
House S — Volume = 203.97 m³, Underground surface area = 124.5 m²					
S-1	886	0.402	24.4	0.355	17.5
S-2	1107	0.531	7.7	0.536	12.1
House W — Volume = 229.46 m³, Underground surface area = 83.6 m²					
W-1	586	0.571	32.4	0.461	59.5
W-2	1050	0.447	34.3	0.748	34.6
W-3	802	1.031	23.8	0.992	17.4
W-4	440	1.329	92.2	1.118	96.9
W-5	961	1.643	15.7	1.664	13.9
W-6	1338	1.161	10.5	1.036	10.3
W-7	1000	1.169	20.6	1.032	21.1
W-8	145	1.552	15.6	2.166	29.6
W-9	330	2.741	3.2	2.851	7.1
W-10	525	1.383	27.9	1.349	30.9
W-11	325	1.255	28.1	1.293	10.7

Net percent standard deviation = 31.1
Without run F-1 = 21.6

*Column 1: run numbers
Column 2: equilibrium, steady-state count rate (count/h)
Column 3: ACH by radon method (h⁻¹)
Column 4: percent standard deviation for radon method
Column 5: ACH by reference method (anemometer or SF₆)
Column 6: percent standard deviation of reference method value.