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# SEASONAL AND ANNUAL AVERAGE RADON LEVELS IN 70 HOUSES

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## ABSTRACT

A year-long, multipollutant, indoor air quality study involving 70 occupied houses in four states was completed in 1987. All of the houses included in the study had a partial or complete basement with a concrete slab floor and block walls. On an approximately quarterly schedule, integrating monitors for short-lived radon progeny, NO2, HCHO, and H2O were exposed for one week in each house on both the basement and main levels.

At the beginning of the study, a pair of alpha track detectors were placed both on top of the refrigerator in the kitchen (or some other sampling site on the main floor) and at a sampling site in the basement. One detector at each sampling site was left in place for a year. The other detector at each sampling site was retrieved after about three month's exposure and replaced with another detector for the next quarter. In addition, short-term samples of radon and radon progeny were made at all sampling sites once per quarter. In this paper, comparisons will be made between: (1) seasonal and annual averages, (2) summer and winter averages, and (3) short and long term measurements.

## INTRODUCTION AND STUDY OVERVIEW

Recently, houses have been discovered with levels of radon[1] and its short-lived progeny in excess of federally mandated occupational exposure limits. Radon in its gaseous state is not thought to be a potent lung carcinogen but is the precursor of short-lived, alpha-emitting progeny which are carcinogenic. Excessive exposure to radon progeny resulted in excess deaths from lung cancer in mining populations (1). Nero et al. (2) have estimated that about one million homes have levels in excess of 300 $Bq/m^3$.[2] The discovery of homes with markedly elevated radon levels has resulted in increased awareness of the need for surveys of radon levels in typical American homes.

An indoor air quality study of seventy houses in four states during 1985-87 has been completed by the staff of Oak Ridge National Laboratory (ORNL) and their collaborators. Table 1 describes the distribution of study houses among seven cities and the schedule of visits to the houses for sampling. Core activities were conducted in each house each season. One-week average levels of the following were determined: radon progeny, nitrogen dioxide, water vapor, formaldehyde, vapor-phase polynuclear aromatic hydrocarbons, and air exchange rate. In addition, instantaneous levels of radon and radon progeny and three-month and twelve-month levels of radon were determined. All of these measurements, except one-week average levels of radon progeny, were generally made at two sampling sites in each house. The "downstairs" site was in the basement and the "upstairs" site was on the floor immediately above (i.e., the main floor).

Supplemental information was gathered in a number of ways. A detailed questionnaire concerning house characteristics and occupant activities was administered to each homeowner. A floor plan of each house was recorded along with a detailed survey of the gamma radiation field in and near the house. A detailed characterization of energy use and weatherization status of each house was performed by utility trained personnel if the homeowner agreed.

Several special studies were part of the overall study. A detailed instrumental characterization of residential microenvironments occurred in six houses during the summer of 1985. A monitoring and data acquisition system was installed and operated for one week in each house. Data were collected from four sampling sites: two upstairs, one downstairs, and outdoors. About every three minutes, acquisition was made of data on levels of sulfur oxides, nitrogen oxides, carbon oxides, ozone, and freon, a tracer for air exchange studies. Another special study involved making tracer decay air exchange measurements in twenty houses, once in the

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[1]All references to radon in this paper refer to the isotope $^{222}Rn$ unless indicated otherwise.
[2]One pCi/L equals 37 $Bq/m^3$.

summer and once in the winter. A radiological survey was made of Madison County, AL, to characterize potential radon sources in that region.

## STUDY DESIGN AND EXPERIMENTAL METHODS

Single family detached dwellings, most with basements, were selected in four states. The study houses were of various ages, styles, and construction materials. Several methods were tried in our efforts to recruit volunteer participants for the study. Initial efforts consisted of obtaining mailing lists from local electric power distributors and sending solicitation letters. There were no participants successfully recruited in this way. There were also no successful participants recruited when the initial contacts were made via telephone. Most of the participants were recruited after a series of talks describing the study were given to academic, industrial and civic groups near the targeted cities (i.e., Chattanooga, Rossville, Birmingham, Huntsville, Florence, and Tupelo). When members of the audience reacted favorably to our request for volunteers, we explained to them that the requirements[1] of the study included presence of block wall construction in the lowest level of studied houses with at least one other higher level and that at least eight visits would be made to each house. If they were still interested, then we encouraged them to discuss the study with their neighbors and encourage a cluster of homes to volunteer. This resulted in a geographical distribution of homes that facilitated the logistical operation of the study. Another method by which interested potential participants were identified was the placement of news articles in newspapers in the targeted cities. People who responded to these articles were also encouraged to discuss the study with their neighbors.

Much use was made of integrating samplers in the study. A photograph of the integrating samplers used is shown in Figure 1. Alpha track monitors were used to measure longer-term average levels of radon in the homes. The monitors were obtained commercially[2] and are functionally similar to those described by Alter and Fliescher (3). Study personnel placed and retrieved the monitors, which were oriented as shown in Figure 1. Monitors were generally placed at the two sampling locations in each house, upstairs and downstairs. Modified thermoluminescent (TLD) dosimeters (referred to as MODs) were used to measure one-week average progeny levels (4). Analysis of the TLD chips was performed at TVA's Western Area Radiological Laboratory (WARL) by C. N. Evans.

Modified Lucas alpha scintillation cells were used to measure short-term average levels of radon. The cells were made at ORNL and are functionally similar to those described by Volchok and dePlanque (5). The cells were counted prior to use to obtain a background count. For each

[1]For the final 10 houses in the Birmingham area the requirements were less restrictive.
[2]Terradex Corporation, 3 Science Road, Glenwood, IL, 60425-1579.

measurement, a 500-mL air sample was drawn into a cell and the cell was held for at least three hours prior to counting. A modified motor home contained the pulse counting system used for the ORNL cells. Figure 2 is a photograph showing the mobile laboratory vehicle parked in front of one of the study houses. Other radiation-induced pulse counting systems and computers were also contained in this vehicle. Each quarter this vehicle visited each of the houses in the study.

Alpha spectroscopic analysis of material collected on 0.45-$\mu$ filters from ten-minute air samples of 100-200 ℓ was used to estimate short-term average levels of $^{218}Po$, $^{214}Bi$, $^{214}Po$ and sometimes $^{212}Pb$ and $^{212}Bi$. The counting system used has been described (6,7) and was contained in the vehicle shown in Figure 2. The results were analyzed with a computer program that makes use of mathematical techniques suggested by Martz et al. (8) and by Duggan (9). Figure 3 is a photograph of the air sampling pump in one of the study homes.

In most cases, one integrating sampler of each type was left at both the upstairs and downstairs sites each quarter. About 5% of the samplers used were deployed in either triplicate or duplicate to estimate precision. About 5% of the samplers were kept at ORNL, never exposed, and returned to the vendor's laboratory for analysis to allow us to estimate analytical background values. Of 24 unexposed alpha track monitors, the mean level was 6.66 Bq/m$^3$ with the standard deviation of the distribution being 3.33 Bq/m$^3$. Data reported here were not corrected for these glank values because the mean blank response is not greater than zero ($P > 0.05$). Variance among the replicate exposures were statistically analyzed in groups determined according to the mean levels detected by sets of replicate monitors. Table 2 summarizes the results. The accuracy of the monitors was determined by the vendor (10). Monitors from two manufacturing lots were exposed under well controlled experimental conditions. On each occasion, exposure occurred in two different radon chambers and calibration coefficients were determined. The relative standard errors for each group of monitors were 2.5% and 2.4% of the mean. Based on these results we estimate that the total error in our radon measurements to be less than ±20% under field conditions.

The cells with which short-term (i.e., nearly instantaneous) measurements of radon were made were calibrated on several occasions. Variation in the calibration factor was typically about ±20%.

About 10% of all MOD placements were utilized to monitor the quality of the integrated measurements under field conditions. On 57 occasions, MODs were deployed in pairs. The variance in the replicated data values was analyzed and the mean and standard error were found to be 325 ± 125 nJ/m$^3$. On 25 occasions, unexposed TLD chips were returned to the laboratory for analysis. The mean and standard deviation of the blanks (based on a nominal exposure time of 168 h and flow of 0.1 L/min) were 0.4 ± 3.6 nJ/m$^3$, which is not significantly different from zero

(P > 0.50)[1]. The accuracy of these measurements was estimated from data obtained in the radon progeny chamber at the U.S. Department of Energy's Environmental Measurements Laboratory (EML) in New York. On two occasions, nine samplers were exposed in sets of three for one, two, or three days in the chamber at EML. On both occasions, three unexposed samplers were sent with the exposed samplers to the laboratory at WARL for thermoluminescent analysis. Experimentally observed results were regressed onto expected (EML) results. The linear regression estimate (± standard error) of the slope was 1.159 ± 0.085 and of the intercept was 3180 ± 22900 (nJ/m$^3$)-h. The slope is not different from unity (P > 0.05) and the intercept is not different from zero (P > 0.05). Based on these results, we estimate the overall error under field conditions to be around ±40% for potential alpha energy concentration (PAEC) levels below 500 nJ/m$^3$ and less than ±20% for higher levels.

In August, 1986, the alpha spectroscopic measurement system used for short-term progeny measurements was taken to the U.S. Environmental Protection Agency's Eastern Environmental Radiation Facility (EERF) in Montgomery, AL, for intercomparison with the EERF measurement system. Twelve sets of measurements were made and good agreement between ORNL and EERF was obtained and we estimate that errors are probably less than 25% for isotopic measurements under field conditions.[2]

## RESULTS

Preliminary comparisons have been made between annual average radon levels and corresponding seasonal average values. The ratio of the measured annual average radon level to the time-weighted average of the four measured seasonal average radon levels ranged from less than 0.5 to over 2.5 (see Figure 4) with typical values being around 0.80. Annual average radon levels were regressed onto the weighted average of four seasonal averages for data from both sampling sites (see top panel of Figure 5). The intercept is not significantly (P > 0.05) different from zero. The slope is significantly (P < 0.05) less than unity. To investigate this finding further, regression analyses were performed on subsets of the data. Because airborne dust levels might be typically higher in basements compared to living areas, regression analyses were performed separately on basement and living-area results that were less than 300 Bq/m$^3$ (see bottom panel of Figure 5). No significant difference (P > 0.05) was found between the basement and living-area regressions. In future work, these data will be further analyzed to elucidate the potential causes for these findings.

Comparisons have been made between annual radon measurements taken in basement and living-area locations. The average and range of annual alpha

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[1]Data reported here were not corrected for these blank values.
[2]Because of the short lifetimes of these isotopes, it was impossible to obtain replicate measurements in the occupied houses.

track measurements in the different cities for measurements taken in the basement and in the living area are summarized in Figure 6. Basement results are consistently higher than living-area results although the difference is not as great as that among cities. Comparisons were made between summer and winter seasonal alpha track measurements. The comparisons for basement and living-area results are summarized in Figure 7. The difference between summer and winter results was found to be small.

Because of the relatively high levels of radon found in the Huntsville houses, we attempted to characterize the source of radon in these houses. Samples of building materials were analyzed by energy dispersive X-ray analysis for indications of Florida phosphate. Samples of building material, soil, and outcropping rocks were analyzed for $^{226}Ra$, $^{232}Th$, and $^{40}K$ using high-resolution gamma spectroscopy. The radium content of the materials in or near these houses did not seem to be great enough to fully account for the radon levels observed. Geologists from ORNL's field office in Grand Junction made a survey trip to Madison County during August, 1986. The radium content of each of the geological strata that outcrop in the county was determined. Chattanooga Shale had a radium content of 30 pCi/g while all other strata had levels less than 2.0 pCi/g. The houses are separated from the Chattanooga Shale layer below them by about 300 feet. The intervening material is known to be quite fractured and it may be possible for a substantial flow of radon-laden gas to move from the shale layer to the surface.

Soil characteristics may also affect radon transport into houses. For each of the houses in the study, soil survey data from the Soil Conservation Service was examined. Three-month average radon levels measured during the first two quarters were compared with characteristics of soil near the house. In a variety of analysis of variance tests, the available data seem to suggest that the drainage and slope of surface soil may significantly be associated with indoor radon levels. This finding is consistent with the fact that houses on or near ridge lines tend to have elevated radon levels compared to neighboring houses in nearby valleys.

Over 400 sets of short-term measurements were made, each of which included nearly simultaneous values of $^{222}Rn$, $^{218}Po$, $^{214}Pb$, and $^{214}Bi$. Levels of $^{218}Po$ were found to be closest to being in secular equilibrium with $^{222}Rn$ (i.e., progeny ratio equals unity), with $^{214}Pb$ and $^{214}Bi$ being increasingly farther from equilibrium. Progeny ratios were not markedly different between basement and living-area locations, suggesting that there was rather rapid transport of radon-laden soil gas from the point of entry (presumably the basement) to the rest of the house. Progeny ratios were also examined on a house-by-house basis (results not shown) and no significant difference was seen between living-area and basement results.

Airborne particulate levels are thought to play a major role in determining the fate of newly generated radon progeny (1,11). In brief, a newly generated progeny is an airborne, ionic species which can undergo one of three outcomes: it can attach to an airborne particle and become part of the attached fraction, it can plate out on a wall or other surface and cease to be airborne, or it can undergo radioactive decay before either of the first two events occur. If the concentration of airborne particles were to be reduced without markedly increasing the air exchange rate, then more progeny would undergo plateout or radioactive decay. Therefore, it is expected that, per unit of $^{222}$Rn activity, levels of $^{218}$Po will be little affected by reduced particulate levels, levels of $^{214}$Pb will be somewhat reduced, and levels of $^{214}$Bi will be markedly reduced.

One of the Knoxville houses (House #942) had an electrostatic precipitator operating within the plenum of the air handling system of the home heating system. The precipitator resulted in much lower levels of airborne respirable particles compared to other houses in the study. For data from the six houses in the vicinity of Knoxville, scatter plots of short-term levels of $^{218}$Po, $^{214}$Pb, and $^{214}$Bi versus simultaneously determined, short-term levels of $^{222}$Rn were examined. In the case of $^{218}$Po, there is no discernible difference in the data trends for House #942 compared to the other houses. In the cases of $^{214}$Pb and $^{214}$Bi, the progeny ratios for House #942 are clearly less than for the other houses, which is consistent with the lower levels of respirable particles found in this house.

## DISCUSSION

The most striking pattern in the radon data is the very large difference between data from different cities. Because of the relatively high levels observed in Huntsville, preliminary results from the first half of the study have been reported previously (12). Preliminary efforts at source characterization have revealed that radium content of geological strata lying above Chattanooga Shale is very low in the Huntsville area. Future work will address the possibility that radon may migrate almost 300 feet in this area to reach the earth's surface.

Another striking pattern is the absence of markedly elevated radon levels in the winter in many of the cities studied. Current federal guidelines for radon surveys encourage homeowners to test their homes in the winter because winter levels are thought to be higher. This may or may not be the case in some parts of the southeastern U.S.

## ACKNOWLEDGMENTS

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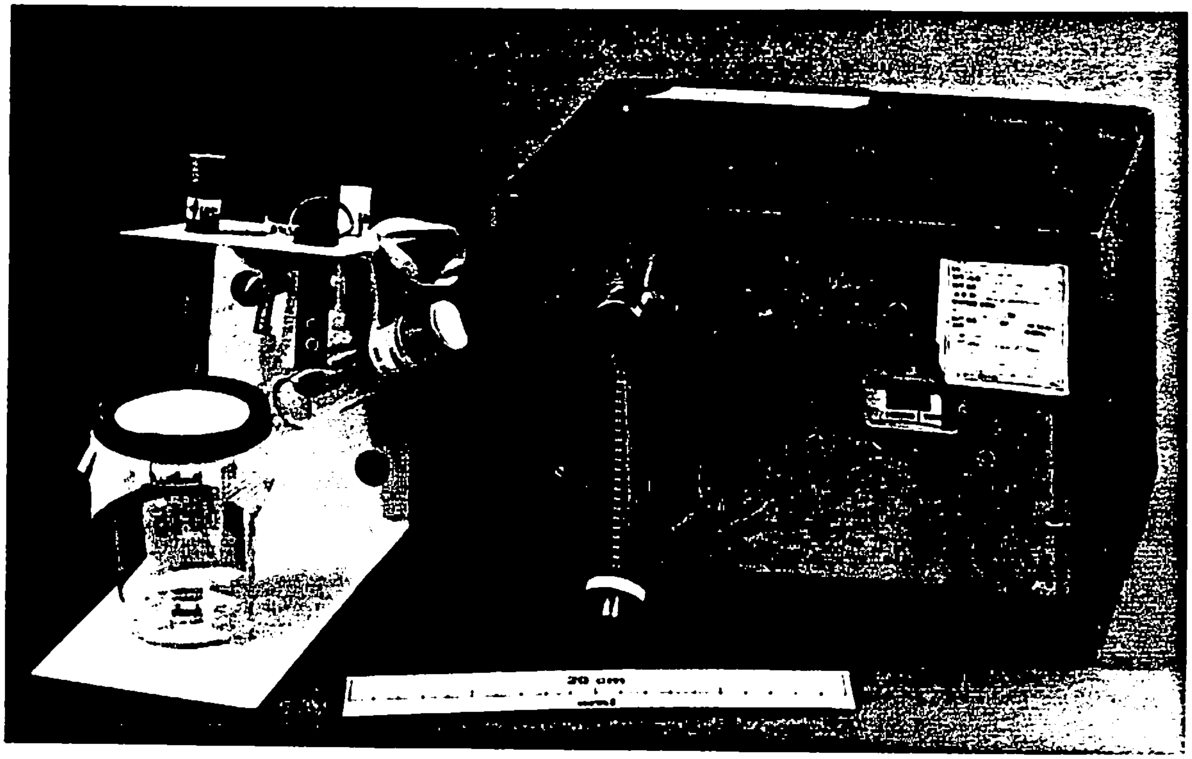

Figure 1. MOD and sampling rack containing passive monitors. (The cup sitting on the rack measures radon. The other passive samplers, from left to right, measure water vapor, perfluorocarbon tracers, polynuclear aromatic vapors, nitrogen dioxide, and formaldehyde. The MOD measures radon progeny.)

Figure 2. Mobile radiological laboratory in front of a house in study.

Figure 3. An air sampling pump collecting airborne ions for spectroscopic analysis of alpha emitters.

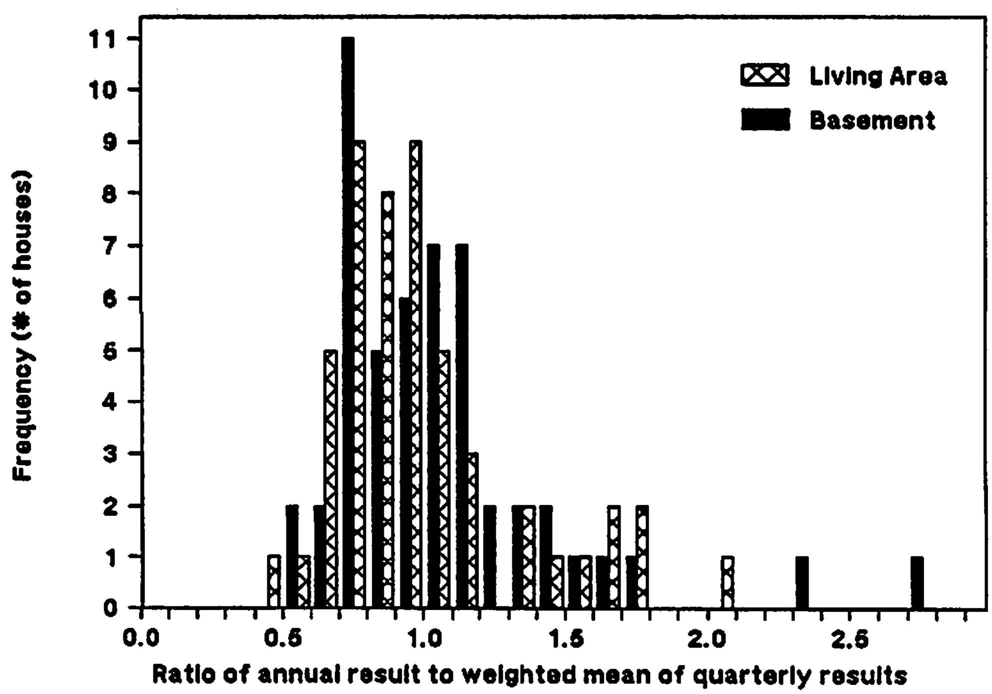


Figure 4. Distribution of ratios of annual radon measurement divided by average of seasonal measurements.

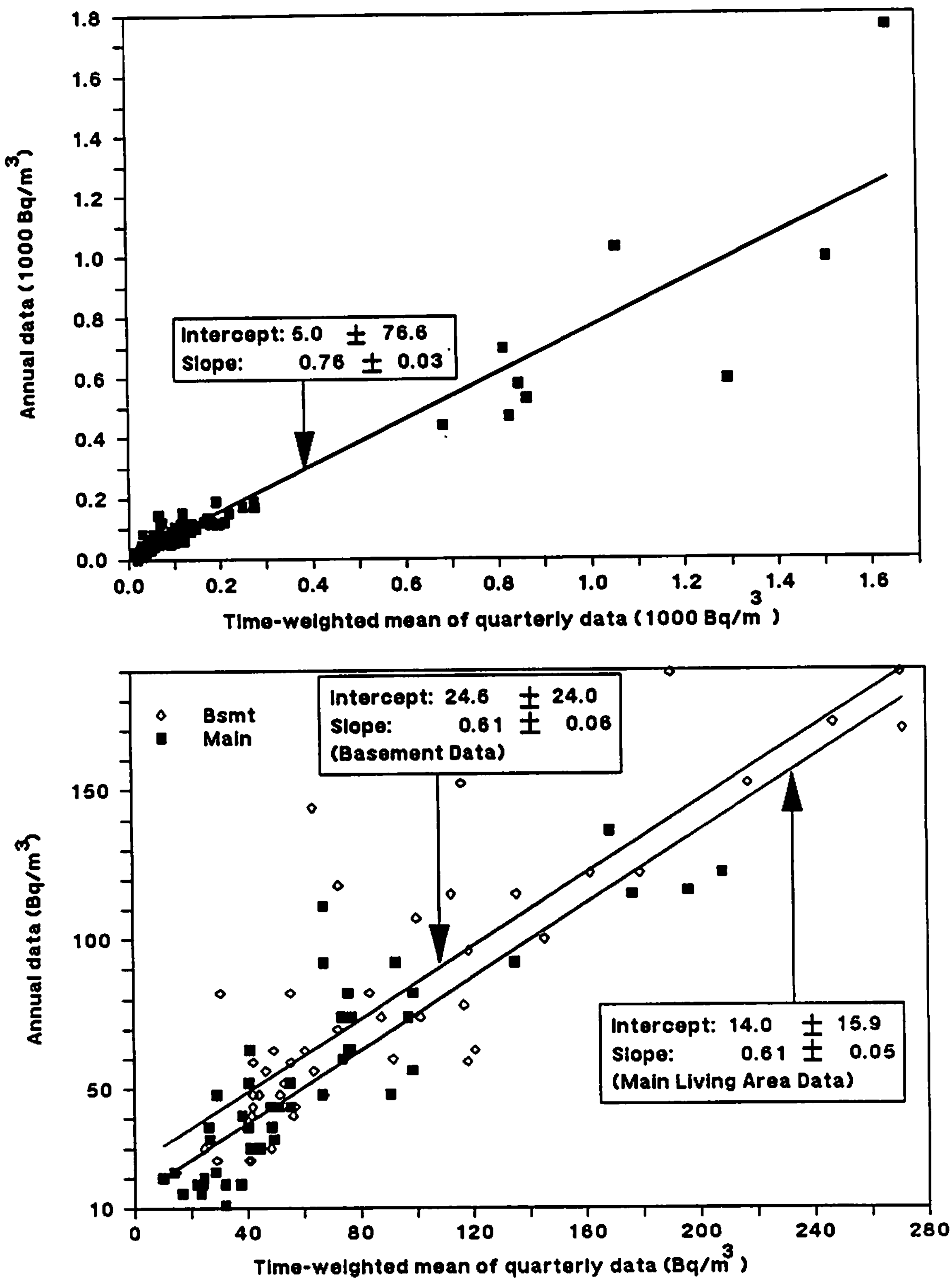


Figure 5. Regression analyses of annual and seasonal (summed) radon measurements.

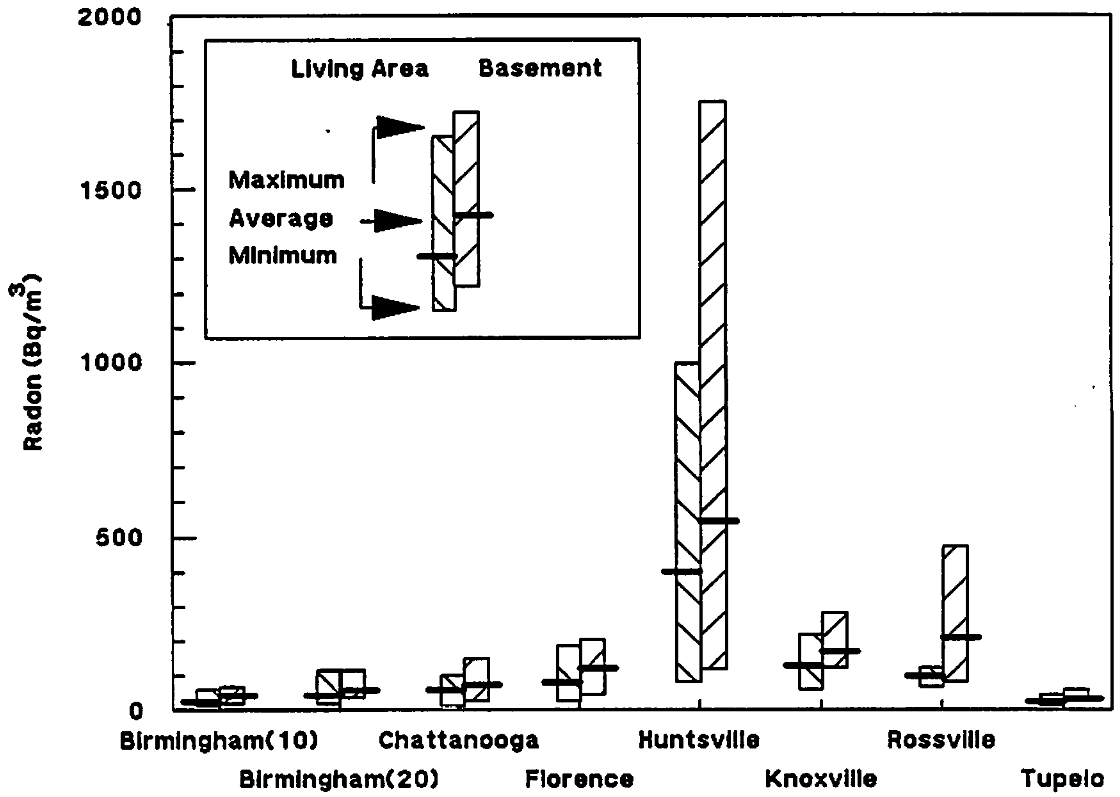


Figure 6. Comparison of annual radon measurements between living-area and basement sites.

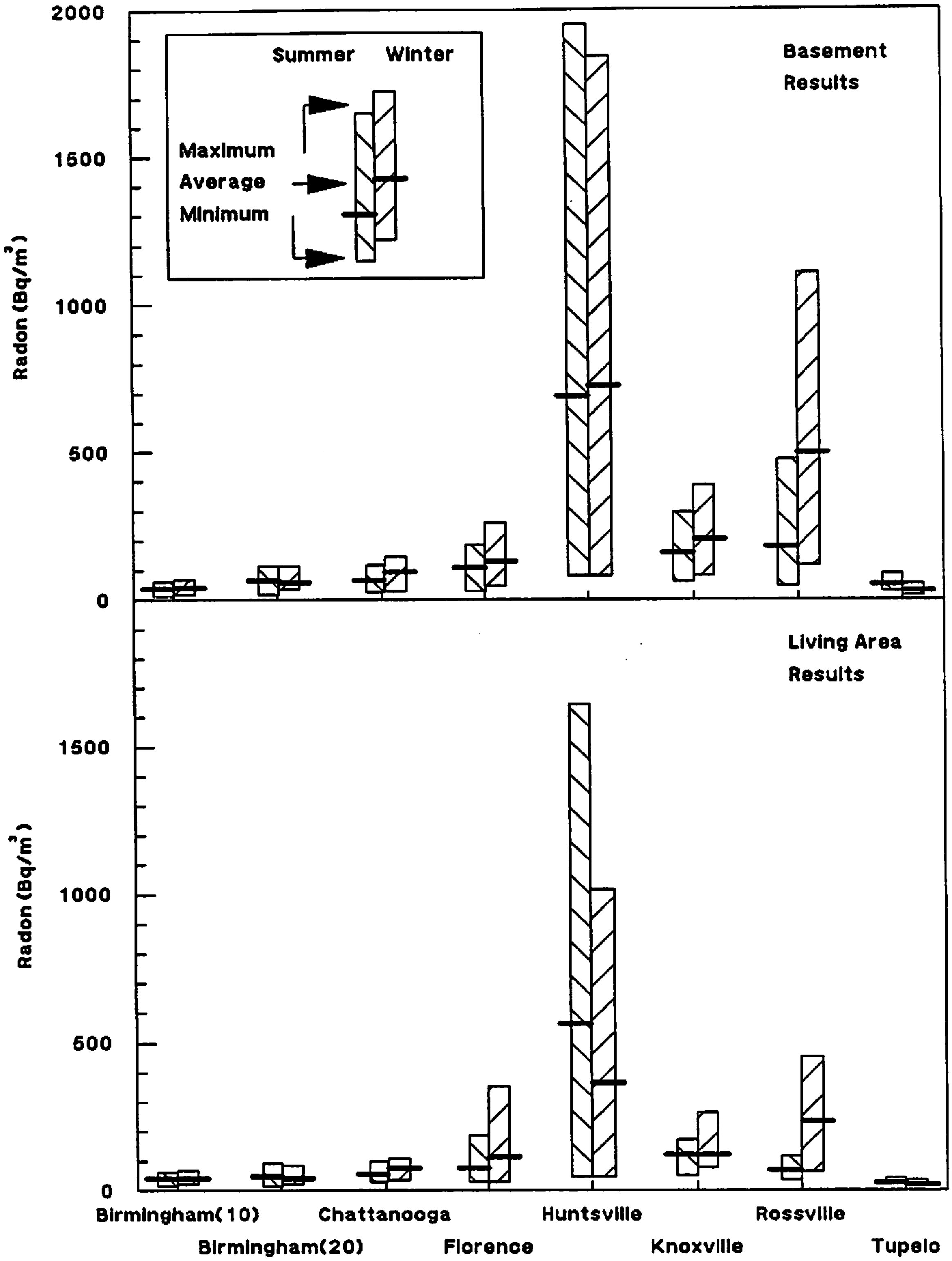


Figure 7. Comparison of summer and winter seasonal radon measurements for basement (top panel) and living-area (lower panel) sites.

TABLE 1. SCHEDULE OF INDOOR AIR QUALITY SAMPLING VISITS TO SOUTHEASTERN CITIES

| City | Number of Houses | Summer Visit | Autumn Visit | Winter Visit | Spring Visit |
|---|---|---|---|---|---|
| Birmingham, AL* | 20 | 8/85 | 10/85 | 1/86 | 4/86 |
| | 10 | 8/86 | 11/86 | 1/86 | 4/86 |
| Huntsville, AL | 8 | 9/85 | 11/85 | 3/86 | 6/86 |
| Florence, AL | 8 | 9/85 | 11/85 | 3/86 | 6/86 |
| Rossville, GA | 4 | 7/85 | 10/85 | 1/86 | 4/86 |
| Tupelo, MS | 4 | 9/85 | 11/85 | 3/86 | 6/86 |
| Chattanooga, TN | 10 | 7/85 | 10/85 | 1/86 | 4/86 |
| Knoxville, TN | 6 | 7-8/85 | 11/85 | 2/86 | 5/86 |

*Ten houses in the Birmingham area joined the study in 1/86.

TABLE 2. ANALYSIS OF REPLICATE RADON DATA

| Range of Replicate Means ($Bq/m^3$) | Mean ($Bq/m^3$) | Standard Deviation | CV (%) | Number of Replicates |
|---|---|---|---|---|
| 20-75 | 50 | 9 | 18 | 15 |
| 75-370 | 145 | 25 | 17 | 24 |
| >370 | 732 | 137 | 19 | 7 |