Table 25.10. Results of in-situ soil measurements at CARL site

Series	Radionuclide	Soil concentration (pCi/cm^3)	Dose rate (microrads/hr)
Uranium	^{214}Pb	2.10 ± 0.21	
	^{214}Bi	1.57 ± 0.09	
	Average	1.65 ± 0.09	1.78 ± 0.10
Thorium	^{208}Tl	1.06 ± 0.10	
	^{212}Pb	1.79 ± 0.19	
	^{228}Ac	1.13 ± 0.13	
	Average	1.19 ± 0.07	2.01 ± 0.12
Potassium	^{40}K	5.63 ± 0.51	0.60 ± 0.05
Cesium	^{137}Cs	3.11 ± 0.26[a]	0.15 ± 0.01
		Total	4.54 ± 0.16

[a]The ^{137}Cs concentrations are in terms of pCi/cm^3 at the surface.

the analysis of the large amount of data generated, a computer program was prepared with the assistance of the Spectrometric Dosimetry Group. Data from the multichannel analyzer on the mobile laboratory are punched onto paper tape. Upon return to ORNL the paper tape is read onto magnetic tape at the computer center. The magnetic tape is recalled at a remote terminal, and a FORTRAN file is written. The FORTRAN program BGAPE.F4 operates on this data file to find peaks, to integrate under the peaks, to assign energies based on calibration information, and to calculate soil concentrations using predetermined detector parameters.

In conclusion, we have developed an in-situ gamma measurement system which can be used conveniently in the field to obtain concentrations and dose rates from radionuclides in the soil. With slight modification, this system can be used for routine measurements in the environmental monitoring program at ORNL.

MEASUREMENTS OF RADON DAUGHTER CONCENTRATIONS IN AIR

An alpha-particle spectrometer developed at ORNL[21] and a spectrometry technique reported by Martz et al.[22] have been used in measurements of the effects of sealants on the emanation of radon from concrete.[23] In the spectrometry technique of Martz et al., the concentrations of the short-lived radon daughters RaA, RaB, and RaC attached to aerosols are calculated from one count rate of the RaA and two count rates of the RaC′ alpha-particle activity collected on a filter with an air-sampling device. Because the RaA alpha particles collected on the filter are counted separately, this technique offers more accuracy in the calculated concentrations, especially those of RaA, than other methods of taking three counts of the combined alpha-particle activity of RaA and RaC′ collected on the filter.[24,25]

In the spectrometer developed at this laboratory, helium is flowed between a silicon diode detector and a filter which are separated by a distance of about 0.5 cm. By using helium, the counter can be operated at atmospheric pressure, with considerable gain in filter-handling simplicity and little loss of resolution compared with measurements of the alpha-particle spectrum of RaA and RaC′ on the filter made in a vacuum. The resolution of the spectrometer for air samples collected at a flow velocity of about 50 cm/sec with a membrane (Metricel, Gelman GN-6) and glass-fiber (Acropor, Gelman AN-450) filter having a 0.45-μ pore size is compared with a more porous filter (Whatman grade 4) in Fig. 25.8. Either of these membrane or glass-fiber filters with a medium pore size of 0.45 to 0.80 μ allows easy resolution of the RaA and RaC′ alpha-particle activity on the filter. At flow velocities of up to 100

21. P. T. Perdue, W. H. Shinpaugh, J. H. Thorngate, and J. A. Auxier, *Health Phys.* 26, 114 (1974).

22. D. E. Martz, D. F. Holleman, D. E. McCurdy, and K. J. Schiager, *Health Phys.* 17, 131 (1969).

23. J. A. Auxier, W. H. Shinpaugh, G. D. Kerr, and D. J. Christian, to be published in *Health Physics.*

24. E. G. Tsivoglou, H. E. Ayer, and D. A. Holaday, *Nucleonics*, 11(9), 40 (1953).

25. J. W. Thomas, *Health Phys.* 23, 783 (1972).

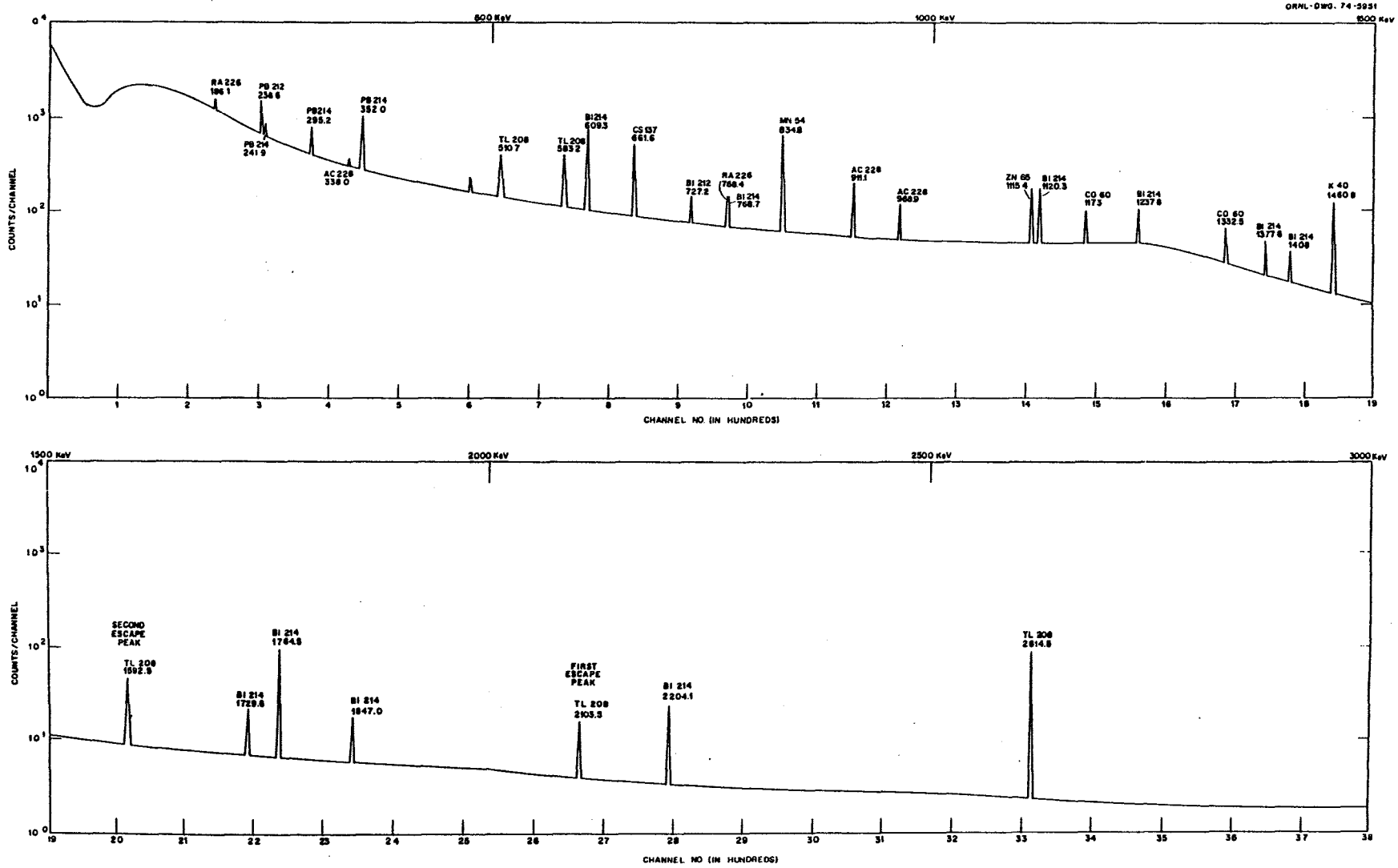

Fig. 25.7. Typical Ge(Li) detector spectrum of gamma rays from natural and fallout radionuclides in soil.

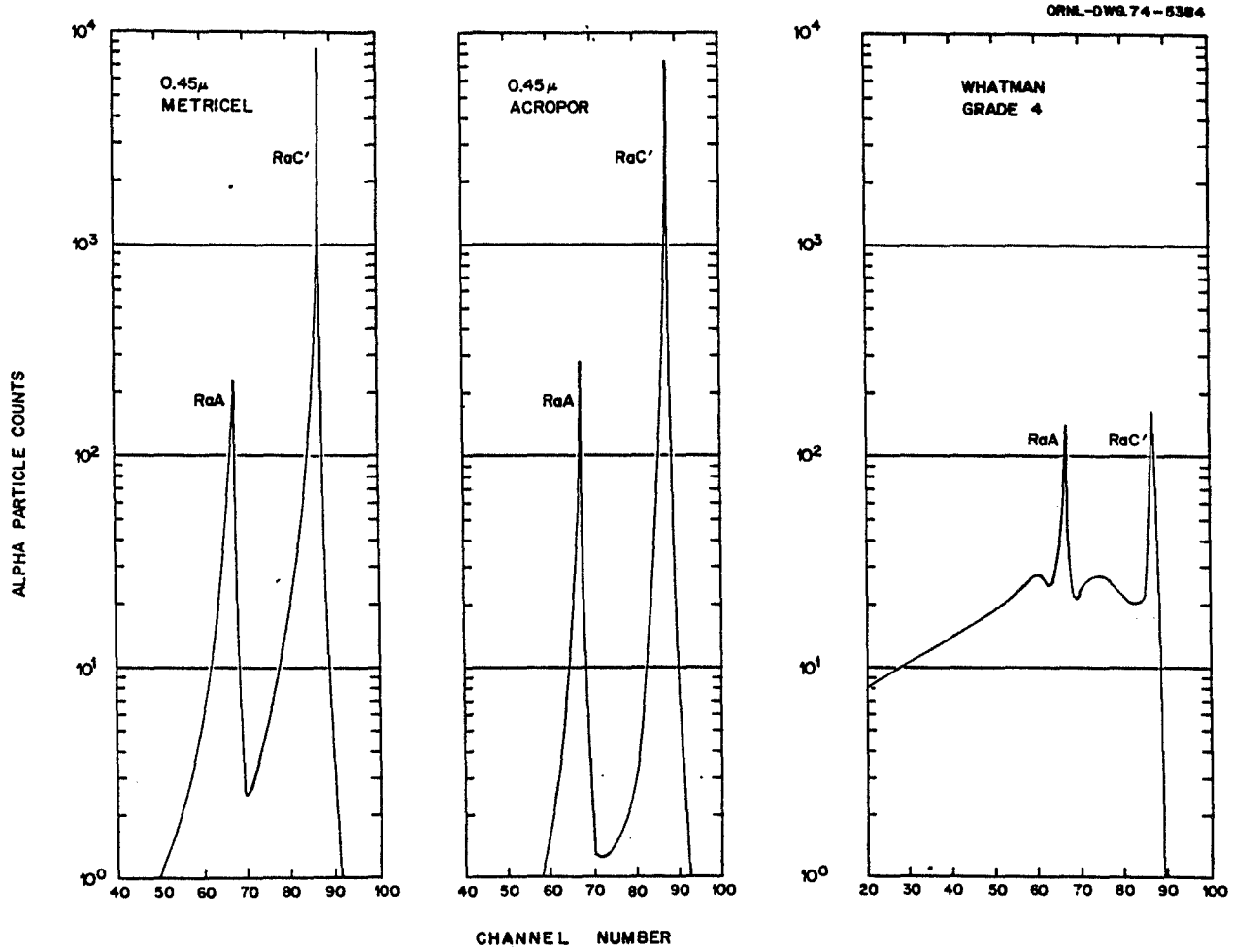

Fig. 25.8. Comparison of the spectra of radon daughter radionuclides collected on various filters and counted with alpha-particle spectrometer described in refs. 17 and 21.

cm/sec, these filters are normally more than 99% efficient in collecting aerosols.[26]

A modification of the spectrometry technique has also been developed at this laboratory[27] to improve the accuracy and sensitivity of radon daughter measurements. In this modification, radon daughter concentrations are calculated from one integral count of the RaA and two integral counts of the RaC′ alpha-particle activity collected on a filter. A computer program, RPCON4, has been written which will calculate the air concentrations and estimate the accuracy in the calculated concentrations of RaA, RaB, and RaC. This program is available in the BASIC language.

For sampling times in the range of 5 to 15 min, one RaA counting interval from 2 to 12 min and two RaC′ counting intervals from 2 to 12 min and from 15 to 30 min after the termination of the air sample collection have been found to give a good overall accuracy in the calculated concentrations for a wide range of RaB/RaA and RaC/RaA activity ratios. The starting time of 2 min for the first is the shortest practicable time for transferring the filter from the air sampling device to the spectrometer, and an ending time of 30 min is standard for techniques of this type.[22,24,25] Air sampling time intervals greater than 15 min were also investigated but were not found to offer any great improvement in the accuracy of the calculated concentrations.

In late October and early November of 1973, the alpha-particle spectrometer and modified spectrometry

26. American National Standards Institute, *Guide to Sampling Airborne Radioactive Materials in Nuclear Facilities*, ANSI-N13.1 (1969).

27. G. D. Kerr, *Trans. Amer. Nucl. Soc.* 17, 541 (1973).

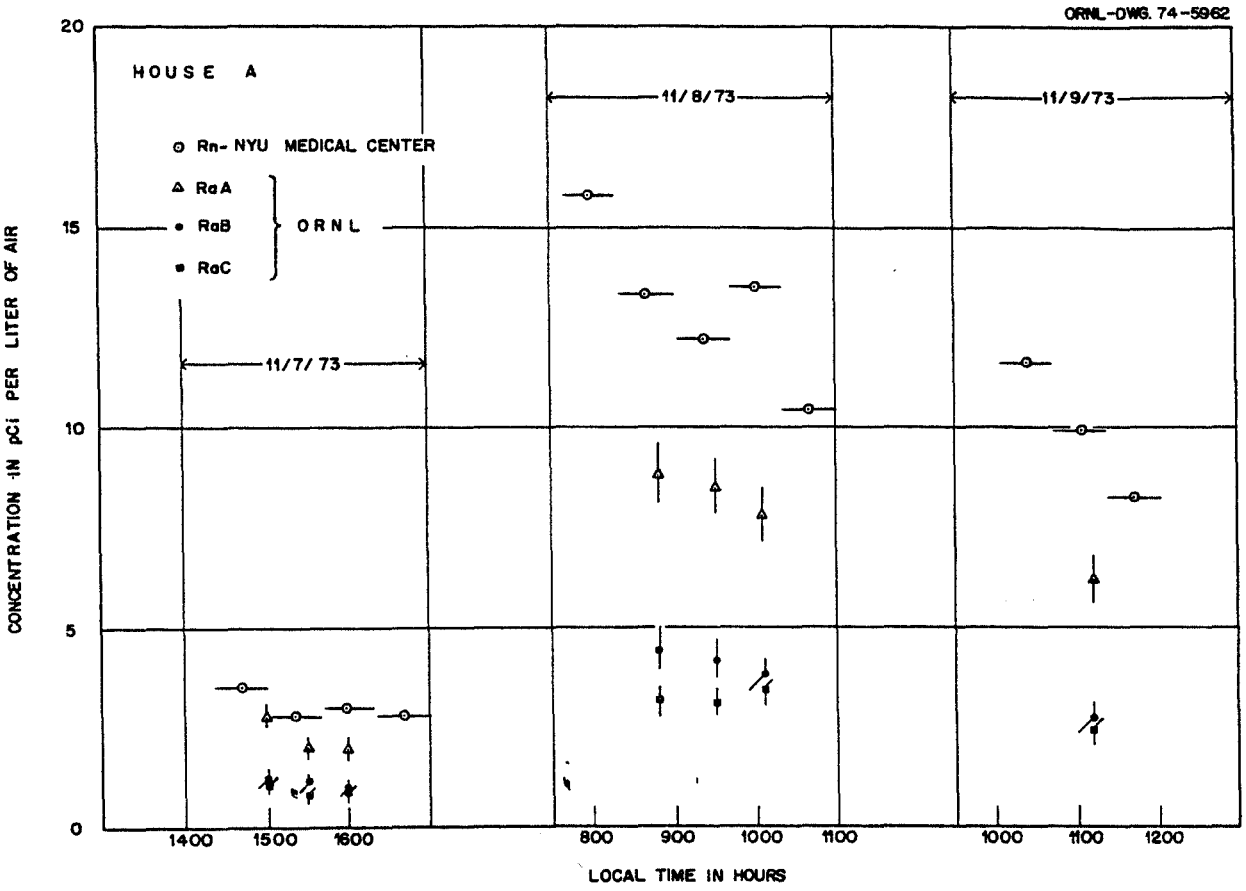

Fig. 25.9. Results of radon and radon daughter measurements in a house at Grand Junction, Colorado, showing variation in concentrations with time.

technique were used to measure radon daughter levels in nine structures in Grand Junction, Colorado, at the request of the AEC. Tailings from uranium mines had been used in their construction, but two had been reconstructed to remove most of the tailings. Because most of the tailings had been used as a fill, the measurements were made in either the basements of the structures or the first floor of slab-type structures. With one exception, the measurements were made in the living areas of the houses. Examples of the radon daughter measurements in one of the structures are shown in Fig. 25.9. Also shown in the figure are measurements of radon levels made by the Institute of Environmental Medicine of the New York University Medical Center.[28] The radon concentrations from their measurements are plotted at the center of 40-min measuring intervals, indicated by the horizontal bars. Radon daughter concentrations from our measurements are plotted at the midpoint of air sampling times of 5 to 15 min.

The vertical bars on the radon daughter concentrations are an estimate of the probable error in the measurements. These are based on a statistical uncertainty of one standard deviation in each of the three counts of activity on the filter and on systematic uncertainties of ±5% in both the detection efficiency of the alpha-particle spectrometer and the air sampling rate. Air sampling rates were measured with a flowmeter that had been calibrated with a wet test meter, using standard procedures.[29,30] Concentrations of

28. H. Spitz and McD. E. Wrenn, personal communications (1974).

29. American National Standards Institute, *Radiation Protection in Uranium Mines and Mills*, ANSI-N7.1a (1969).

30. R. L. Rock, R. W. Dalzell, and E. J. Harris, *Controlling Employee Exposure to Alpha Radiation in Underground Uranium Mines*, U.S. Department of the Interior, Bureau of Mines (1971).

Table 25.11. Average and standard deviation of radon daughter concentrations measured in structures at Grand Junction, Colorado

Structure	Period	Number of measurements	Average concentration (pCi/liter)			Trend in radon concentration
			RaA	RaB	RaC	
House A	Afternoon	3	23 ± 0.5	1.1 ± 0.2	1.0 ± 0.2	Falling slowly
	Morning	4	7.9 ± 1.2	3.8 ± 0.8	3.0 ± 0.4	Falling
House B	Morning	2	40 ± 5	25 ± 6	19 ± 6	Falling
	Afternoon	2	25 ± 5	19 ± 4	18 ± 3	Falling slowly
House C	Afternoon	3	0.8 ± 0.3	0.2 ± 0.04	0.4 ± 0.08	
	Morning	2	1.4 ± 0.4	0.6 ± 0.2	0.5 ± 0.08	
House D	Afternoon	3	28 ± 3	11 ± 0.9	8.4 ± 1.2	
	Morning	2	21 ± 11	9.1 ± 2.6	4.6 ± 1.8	Falling
House E	Morning	2	10 ± 4	5.5 ± 2.0	4.6 ± 1.2	Falling
House F	Afternoon	5	232 ± 22	172 ± 17	145 ± 14	Steady
House G						
Location 1	Afternoon	2	28 ± 5	13.2 ± 0.2	8.2 ± 0.1	Falling slowly
	Morning	1	38	15	11	Rising
Location 2	Afternoon	2	23 ± 7	13 ± 2	6.8 ± 0.4	Falling slowly
	Morning	1	42	14	9.2	Rising
House H	Afternoon	4	5.3 ± 0.4	3.1 ± 0.2	2.0 ± 0.3	Steady
	Morning	2	6.0 ± 0.7	3.4 ± 0.4	2.0 ± 0.6	Steady
School	Afternoon	3	0.8 ± 0.4	0.2 ± 0.09	0.2 ± 0.1	Rising slowly

RaA, RaB, and RaC measured in these structures varied from highs of 255 (±7%), 187 (±8%), and 156 (±7%) pCi/liter, respectively, in a nonreconstructed house to lows of 0.50 (±20%), 0.17 (±35%), and 0.15 (±30%) pCi/liter, respectively, in a school.

Because the general pattern of our measurements was to move from one structure in the forenoon and to set up at the next structure in the afternoon, the late afternoon and early morning measurements, which showed considerable variation in some structures, were averaged separately. The average and the standard deviation of the measured radon daughter concentrations for each set of afternoon or morning measurements are given in Table 25.11. Based on the NYU Medical Center measurements, trends in the radon levels spanning the time interval of our measurements in the structures were determined and are shown in Tables 25.11 and 25.12. In Table 25.12 the averages and standard deviations of the RaB/RaA and RaC/RaA activity ratios are given for each set of corresponding measurements in Table 25.11. The average activity ratios in Table 25.12 show that the closest approaches to an equilibrium condition (i.e., RaB/RaA = RaC/RaA = 1) were in the afternoon measurements at house B (RaB/RaA = 0.76 and RaC/RaA = 0.74) and house F (RaB/RaA = 0.74 and RaC/RaA = 0.63).

Large fractional standard deviations in the concentrations or the activity ratios of Tables 25.11 and 25.12, respectively, are mainly associated with either rapid changes in the radon levels or large uncertainties in the alpha-particle counts at the lower radon daughter levels. At higher and either steady or slowly changing radon daughter levels, very good estimates of the ventilation rate of a structure could be made from the average activity ratios and the steady-state solutions to the ventilation equations of Hultiqvist.[31] In five structures where estimates could be made within one standard deviation of the activity ratios, the ventilation rates ranged from 0.4 to 1.6 air exchanges per hour.

The ventilation rates of structures are, of course, affected by the type of heating system used, the opening of doors, etc., and therefore do not remain constant, as shown by the varying radon and radon daughter levels in Fig. 25.9. However, the range of ventilation rates estimated from the radon activity ratios in these structures compares very well with the range of ventilation rates measured in houses under similar conditions by other methods.[32] During our measurements, most of the houses were closed, and the heating systems were in operation.

31. B. Hultiqvist, *Studies on Naturally Occurring Ionizing Radiations*, Almqvist and Wiksells Boktryskeri AB, Stockholm, 1956.

32. T. H. Handley and C. J. Barton, *Home Ventilation Rates: A Literature Study*, ORNL-TM-4318 (September 1973).

Table 25.12. Average and standard deviation of the activity ratios of radon daughters measured in structures at Grand Junction, Colorado

Ventilation rates are based on activity ratios and steady-state solution to Hultqvist's ventilation equations

Structure	Period	Number of measurements	Average activity ratio		Ventilation rate (air exchanges/hr)	Trend in radon concentration
			RaB/RaA	RaC/RaA		
House A	Afternoon	3	0.48 ± 0.08	0.44 ± 0.06	1.2	Falling slowly
	Morning	4	0.48 ± 0.03	0.38 ± 0.03		Falling
House B	Morning	2	0.62 ± 0.06	0.46 ± 0.10		Falling
	Afternoon	2	0.76 ± 0.01	0.74 ± 0.02	0.4	Falling slowly
House C	Afternoon	3	0.31 ± 0.06	0.52 ± 0.01		
	Morning	2	0.42 ± 0.11	0.33 ± 0.01		
House D	Afternoon	3	0.40 ± 0.01	0.30 ± 0.02		
	Morning	2	0.46 ± 0.11	0.22 ± 0.03		Falling
House E	Morning	2	0.55 ± 0.03	0.47 ± 0.04		Falling
House F	Afternoon	5	0.74 ± 0.03	0.63 ± 0.04	0.5	Steady
House G						
Location 1	Afternoon	2	0.47 ± 0.07	0.30 ± 0.05	1.6	Falling slowly
	Morning	1	0.41	0.29		Rising
Location 2	Afternoon	2	0.59 ± 0.24	0.30 ± 0.10		Falling
	Morning	1	0.35	0.22		Rising
House H	Afternoon	4	0.60 ± 0.08	0.39 ± 0.04	1.3	Steady
	Morning	2	0.57 ± 0.04	0.32 ± 0.06	1.1	Steady
School	Afternoon	3	0.30 ± 0.05	0.32 ± 0.02		Rising slowly

Our laboratory finding that epoxy paint was effective in reducing radon emanation from concrete surfaces has also been verified by the NYU Medical Center[28] measurements of radon levels in some houses at Grand Junction where this technique was used. In our laboratory study, radon emanation from concrete was found to be reduced by a factor of 4 after the application of a heavy coat of epoxy paint.

SHIELDING MATERIALS FOR HIGH-YIELD NEUTRON SOURCE CARRIERS

Several materials have been investigated as suitable replacements for paraffin and fiberboard in source carriers for sealed isotopic neutron sources. Paraffin is, of course, an effective shielding material but does not meet newer impact and fire-resistance requirements of source carriers. Fiberboard meets these requirements, but it is one of the poorer shielding materials for high-yield neutron sources.

Machinable composition materials that are fire and impact resistant and that are suitable shielding materials are Bakelite and Transite. For transporting sources, Bakelite is more suitable as a replacement for paraffin because of its lower density. One disadvantage is, of course, the cost of machining slabs of either material to fit into the steel outer drum of a source carrier or other containers.

A search for a conventional formable material failed to produce any of sufficiently low density that were both fire and impact resistant. One has, however, been developed that appears to be promising as a replacement for paraffin. This material uses polyethylene and boric acid as the primary shielding material with a fire-resistant mineral binder containing fibrous strengthening agents. The material was prepared from a mixture of 20 lb of granulated polyethylene (Hifax type 1900), 4 lb of boric acid (anhydride), 8 lb of barium sulfate (anhydrous), 5 lb of sodium silicate (water glass), 2 lb of asbestos fibers, 1 lb of glass-wool fibers, 90 cm^3 of a surfactant (Du Pont Triton X-100), and 6500 cm^3 of water. This mixture was cast in a 14-in.-diam steel drum with a 2-in.-diam central bore for accepting a neutron source and was slowly cured in a stepwise manner to a final temperature of 140°C.

In Table 25.13, maximum dose-equivalent rates in millirems per hour from an americium-beryllium neutron source in a source carrier made from this material are compared with those from the bare source and from the source in identical source carriers made with several conventional shielding materials. All measurements were made perpendicular to the axis of the drum along