Conf-761071--1

MEASUREMENTS OF RADON DAUGHTER CONCENTRATIONS IN STRUCTURES BUILT ON OR NEAR URANIUM MINE TAILINGS*

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ABSTRACT

A technique is discussed that has been used to measure air concentrations of short-lived daughters of ^{222}Rn in residential and commercial structures built on or near uranium mill tailings in the western part of the United States. In this technique, the concentrations of RaA, RaB, and RaC are calculated from one integral count of the RaA and two integral counts of the RaC' alpha-particle activity collected on a filter with an air sampling device. A computer program is available to calculate the concentrations of RaA, RaB, and RaC in air and to estimate the accuracy in these calculated concentrations. This program is written in the BASIC language.

Also discussed in this paper are the alpha-particle spectrometer used to count activity on the air filters and the results of our radon daughter measurements in Colorado, Utah, and New Mexico. These results and results of other measurements discussed in a companion paper are now being used in a comprehensive study of potential radiation exposures to the public from uranium mill tailing piles.

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*Research sponsored by the US Energy Research and Development Administration under contract with Union Carbide Corporation.

INTRODUCTION

Several processes exist for the recovery of naturally occurring uranium. Generally, ore (whose uranium content by weight ranges from a few tenths of one percent to several percent) is transported from mine to mill where it is crushed and leached. The waste material (mill tailings) from this operation is discarded on mill property and is nearly the same volume and weight as the crushed ore entering the mill. The leaching procedure is specific for uranium; therefore, most of the daughter radionuclides including ^{230}Th, ^{226}Ra, and ^{210}Pb remain in the wastes and are left behind once the facility becomes inactive. Uranium mill tailings are sandy in nature, having a particle size which ranges from about -10 mesh (sand) to -200 mesh (slimes). The concentration of ^{226}Ra is highest in the slime fraction, frequently as high as 3000 pCi/g. In the sand fraction, a concentration of 100-300 pCi/g is common. Radon emanates readily from the tailings, and if the tailings are in intimate contact with a structure or stored nearby, radon is likely to accumulate on the inside. The level of accumulation depends in part on the concentration of ^{226}Ra in the tailings, the total amount of tailings involved, and their specific location with respect to the structure.

Leaching operations have ceased at a number of mills in the western part of the United States. At several of these sites, tailings have been removed for private as well as public use. Examples of this practice include the construction of commercial buildings on property where tailings were pushed back to make way for the buildings, but were not removed from the ground on which the structures were built, and the use of tailings as a stabilizing material under the floor of new residences or as backfill around basement walls. Because of this practice, people working in the commercial buildings, and those living in residences are subjected to elevated radon daughter concentrations ranging from small fractions of normal background levels to more than one working level* (WL).

The radon daughter measurements discussed in this report were made in Colorado, Utah, and New Mexico. Most of these measurements were made in cases where the proximity of tailings to a structure was well known (in intimate contact with a structure or stored on nearby property). Results of these measurements and others discussed in a companion paper are now being used in a comprehensive study of potential radiation exposures to the public from uranium mill tailings. The evaluation of potential exposures to radon daughters inside structures presents a formidable problem to the dosimetrist.

MEASUREMENT METHODS

Numerous techniques have been presented for the measurement of radon daughters.[1,2,3] The measurement philosophy at ORNL centers around the sampling of aerosols in air and the use of alpha spectroscopy techniques for determining the concentration of radon daughters.

A modification of the Martz spectroscopy technique[4] for measuring radon daughters has been developed at this laboratory[5] 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, is

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*One working level is equivalent to 100 pCi/l radon in full equilibrium with its short lived daughters, or any concentration of daughters such that the total potential α energy is 1.3×10^5 Mev.

available which will handle differing air sampling rates, sampling and counting times, and counter efficiencies. This BASIC-language program computes the air concentrations of RaA, RaB, and RaC and estimates the accuracy of these calculated concentrations.[6]

In the spectrometer shown in Figure 1, helium is flowed between a silicon diode detector and a filter which are sepa ted by a distance of about 0.5 cm.[7] By using helium, the counter can be operated at atmospheric pressure with considerable gain in filter handling simplicity and very little loss of resolution compared to alpha-particle counts made in a vacuum (see Figure 2). 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 Figure 3. 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 cm/sec, these filters are normally more than 99% efficient in collecting aerosols.[8]

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 count 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.[4,9,10] 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.

APPLICATION AND RESULTS

In late October and November of 1973, the alpha-particle spectrometer and modified spectrometry technique were used to measure radon daughter levels in nine structures in Grand Junction, Colorado, at the request of ERDA (then AEC). Tailings from a nearby uranium mill had been used in their construction, but two had undergone remedial action to remove most of the tailings. Because tailings had been used as a fill, the measurements were made in either the basements of the structures or the first floor of slab-on-grade 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 Figure 4. Also shown in the figure are measurements of radon levels made by the Institute of Environmental Medicine of the New York University Medical Center.[11] 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 systematic uncertainties of $\pm$ 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.[12,13] Concentrations of RaA, RaB, and RaC measured in these structures varied from highs of 235 (± 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.

Average values for radon daughter concentrations during morning and afternoon periods in some structures in Grand Junction, Colorado, are given in Table 1. The ventilation rates, and therefore the radon and radon daughter levels, of the structures are affected by the types of heating and cooling systems, the opening and closing of doors, etc. Results have been published of some of the New York University Medical Center measurements of the diurnal variations of ^{222}Rn in these and other structures in Grand Junction.[14] Based on their measurements, trends in radon levels spanning the time intervals of our radon daughter measurements in the structures are also indicated in Table 1. Large standard deviations in the average values in Table 1 may be attributed to either rapid changes in radon levels within the structures or large uncertainties in the measurements at the lower radon daughter levels.

With two counting intervals of 2 to 12 and 15 to 30 min used in this work, it is possible to measure equilibrium concentrations of 1 pCi/l each of RaA, RaB, and RaC with relative standard deviations of about 15, 18, and 13 percent, respectively. This assumes an air sampling time of 10 min, an air sampling rate of 10 l/min, and a counter efficiency of 0.20. For these same typical sampling and counting conditions, disequilibrium concentrations of 1, 0.4, and 0.2 pCi/l of RaA, RaB, and RaC can be measured with relative standard deviations of about 15, 26, and 36 percent. If the air sampling time is increased to 15 min and the air sampling rate to 17 l/min, the disequilibrium concentrations of 1, 0.4, and 0.2 pCi/l of RaA, RaB, and RaC can be measured with relative standard deviations of about 11, 18, and 26 percent and the equilibrium concentrations of 1 pCi/l each of RaA, RaB, and RaC can be measured with relative standard deviations of about 11, 12, and 10 percent, respectively.

In 1975 and 1976, this sampling technique was utilized in radiological surveys in the vicinity of a number of inactive uranium mills in western states. Sampling in these instances was limited to periodic spot or grab samples because of the requirement that the equipment be manned at all times and because only one sampler was available. The objective of this sampling program was directed at the determination of the concentration of radon and its daughters for situations where the proximity of tailings to a structure was well known (in intimate contact with a structure or stored on nearby property).

A summary of some radon daughter measurements which were made in Salt Lake City are presented in Table 2. Attention is called to three cases in particular, Buildings 2-4. Building (2) is a sewage treatment plant facility located on property surrounded by the tailings piles from the inactive Vitro Mill. Over the period of these measurements, the working level concentration ranged from 0.003 to 0.02 WL. This room is occupied for short periods each day. Buildings 3 and 4 are commercial structures situated near the boundary of the original tailings pond. Both buildings are constructed over tailings which range to five feet deep and which contain up to 900 pCi/g ^{226}Ra. The resulting working level concentration is highest in the store room (seldom occupied for long periods) of building 3, reaching a level of 1.77. In the shop of building 4, the occupancy is higher and the number of working levels ranges from 0.01 to 0.32. It is noted here that these measurements were made during a period when the temperature was moderate and doors were open frequently. When the building is closed thereby reducing the ventilation rate, the number of working levels will increase. Building 7 is constructed in the same area as 3 and 4, but there are no tailings under the floor. The source of elevated radon levels in this building is the large Vitro tailings pile located at the rear of the building.

During a radiological survey of the inactive uranium mill at Shiprock, New Mexico, hourly measurements of the concentration of radon and progeny were made inside a building now used to house a school for teaching construction equipment skills over a period of 21 hours. The results of this series are presented in Figure 5. Although this building is close to the tailings piles associated with this mill, none are in intimate contact with the structure. Therefore, radon enters the building through ventilation. The maximum working level concentration was 0.021 at 12:15 am. It is noted that after that hour, the total daughter concentration decreases more rapidly than does radon. A probable explanation for this is the decrease in condensable nuclei as dust settles due to limited movement of air.

CONCLUSIONS

A modified alpha spectroscopy technique has been used for the measurement of radon daughters collected on a high efficiency air filter. The concentration of RaA, RaB, and RaC may be estimated by solving a series of differential equations which describe the rate at which radon daughters are collected on a filter. Two counting intervals are used providing one measurement of RaA and two measurements, separated by a known interval, of RaC', actually RaC due to the almost instantaneous alpha decay of RaC'. It was observed that with this technique, concentrations of 1, 0.4, and 0.2 pCi/l of RaA, RaB, and RaC can be measured with relative standard deviations of about 15, 26, and 36 percent. One limitation of the sampling and counting equipment used in this work is that it is not automatic. It is necessary that the filters and counter be handled manually.

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Table 1. Average and Standard Deviation of Radon Daughter Concentrations Measured in Structures at Grand Junction, Colorado

Structure	Period	No. of Meas.	Average Concentration (pCi/liter) RaA	RaB	RaC	Average Working Level (WL)	Trend in Radon Concentration
House A	AM	4	7.9 ± 1.2	3.8 ± 0.8	3.0 ± 0.4	0.038	Falling
	PM	3	2.3 ± 0.5	1.1 ± 0.2	1.0 ± 0.2	0.011	Falling slowly
House B	AM	2	40 ± 5	25 ± 6	19 ± 6	0.24	Falling
	PM	2	25 ± 5	19 ± 4	18 ± 3	0.19	Falling slowly
House C	AM	2	1.4 ± 0.4	0.6 ± 0.2	0.5 ± 0.08	0.0062	
	PM	3	0.8 ± 0.3	0.2 ± 0.04	0.4 ± 0.08	0.0035	
House D	AM	2	21 ± 11	9.1 ± 2.6	4.6 ± 1.8	0.085	Falling
	PM	3	28 ± 3	11 ± 0.9	8.4 ± 1.2	0.12	
House E	AM	2	10 ± 4	5.5 ± 2.0	4.6 ± 1.2	0.055	Falling
House F	PM	5	232 ± 22	172 ± 17	145 ±14	1.65	Steady
House G							
Location 1	AM	1	38	15	11	0.16	Rising
	PM	2	28 ± 5	13 ± 0.2	8.2 ± 0.1	0.13	Falling slowly
Location 2	AM	1	42	14	9.2	0.15	Rising
	PM	2	23 ± 7	13 ± 2	6.8 ± 0.4	0.11	Falling slowly
House H	AM	2	6.0 ± 0.7	3.4 ± 0.4	2.0 ± 0.6	0.031	Steady
	PM	4	5.3 ± 0.4	3.1 ± 0.2	2.0 ± 0.3	0.029	Steady
School	PM	3	0.8 ± 0.4	0.2 ± 0.09	0.2 ± 0.1	0.0027	Rising slowly

Table 2. Radon and Radon Daughters Concentrations Measured in Structures at Salt Lake City, Utah

Date 1975	Time	Location	Concentration (pCi/Liter) Rn	RaA	RaB	RaC	Working Level (WL)
9-25	11:00	Building (1), Outside		0.43 (19)*	0.23 (36)*	0.17 (34)*	0.0022
9-26	10:40	"		0.96 (14)	0.17 (75)	0.46 (20)	0.0040
9-26	14:40	Building (2), SW Room		0.78 (15)	0.52 (26)	0.26 (35)	0.0044
9-26	14:40	"		0.60 (17)	0.30 (34)	0.19 (35)	0.0028
10-2	09:35	"		3.2 (9)	2.8 (23)	2.1 (20)	0.025
10-2	11:35	"		2.3 (10)	1.8 (25)	1.7 (19)	0.018
10-2	16:10	"		1.0 (13)	0.5 (34)	0.6 (21)	0.0061
9-26	11:25	Building (3), Store Room	195.	132. (6)	77. (21)	63. (5)	0.76
10-1	09:43	"		139. (6)	108. (19)	61. (22)	0.92
10-1	16:05	"	557.	372. (6)	179. (22)	130. (20)	1.77
9-26	13:00	Building (4), Shop	76.8	49.1 (6)	32.8 (23)	29.1 (18)	0.32
9-26	13:40	"		16.5 (7)	6.7 (25)	5.9 (19)	0.073[a]
10-1	10:22	"		2.2 (10)	0.4 (38)	0.4 (27)	0.0057[b]
10-1	11:22	"	6.9	4.9 (8)	0.9 (27)	0.4 (40)	0.011[b]
10-1	14:50	"	16.2	9.5 (7)	1.8 (24)	0.8 (34)	0.021[b]
10-1	15:30	"		6.4 (8)	1.2 (26)	0.6 (34)	0.015[b]
10-1	17:10	"	81.3	43.6 (6)	11.3 (20)	4.2 (32)	0.12[c]
9-26	16:50	Building (5), Inside		0.28 (24)	0.06 (93)	0.11 (34)	0.001
9-26	17:30	" , Outside		0.34 (23)	0.10 (46)	0.05 (77)	0.001
9-26	18:35	Building (6), Front Room		1.54 (11)	0.49 (31)	0.36 (28)	0.0054
9-27	09:00	Building (7), Store Room		13.2 (7)	9.8 (22)	7.8 (19)	0.092
9-27	10:45	"		7.7 (7)	5.2 (28)	6.2 (16)	0.057
9-27	11:20	" , Outside		2.1 (10)	1.7 (24)	1.3 (21)	0.016
9-29	12:15	Building (8), Outside		0.68 (16)	0.02(283)	0.19 (26)	0.0015
9-29	14:10	Building (9), S. Basement		0.38 (21)	0.09 (68)	0.14 (33)	0.0014
9-29	14:50	"		0.61 (17)	0.06(107)	0.17 (29)	0.0015
9-29	15:25	" , Outside Roof		0.4 (21)		0.1 (26)	0.0005
9-29	16.45	Building (10), Warehouse	6.6	0.9 (14)	0.7 (28)	0.6 (23)	0.0069[d]
9-29	17:20	"		0.8 (16)	0.6 (30)	0.5 (24)	0.0057

*Probable Error in Percent.
aDoor opened approximately one hour prior to measurement.
bDoors opened.
cSample collected 30 minutes after shop closed.
dNear open door.

LIST OF FIGURES

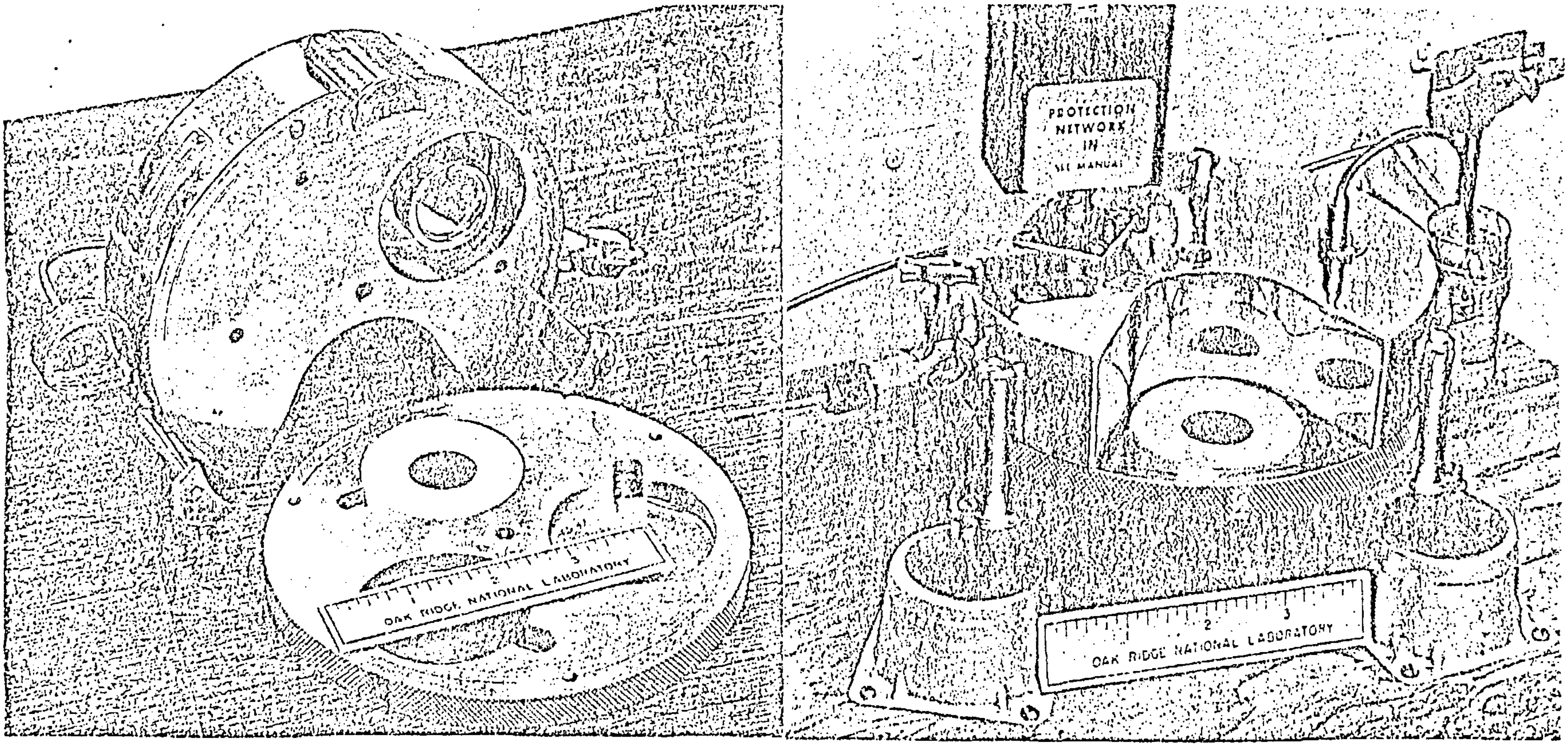
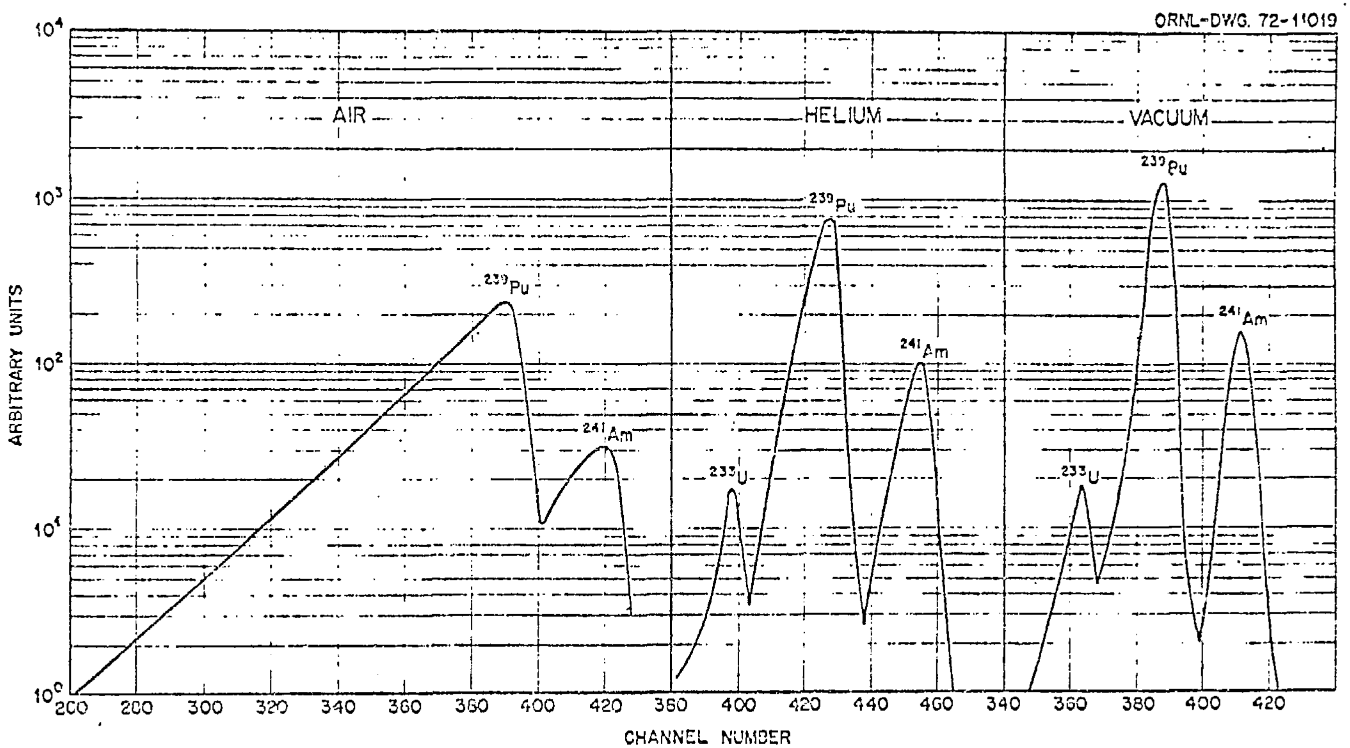

OAK RIDGE NATIONAL LABORATORY
PROTECTION
NETWORK
IN
SEE MANUAL
OAK RIDGE NATIONAL LABORATORY

Fig. 1



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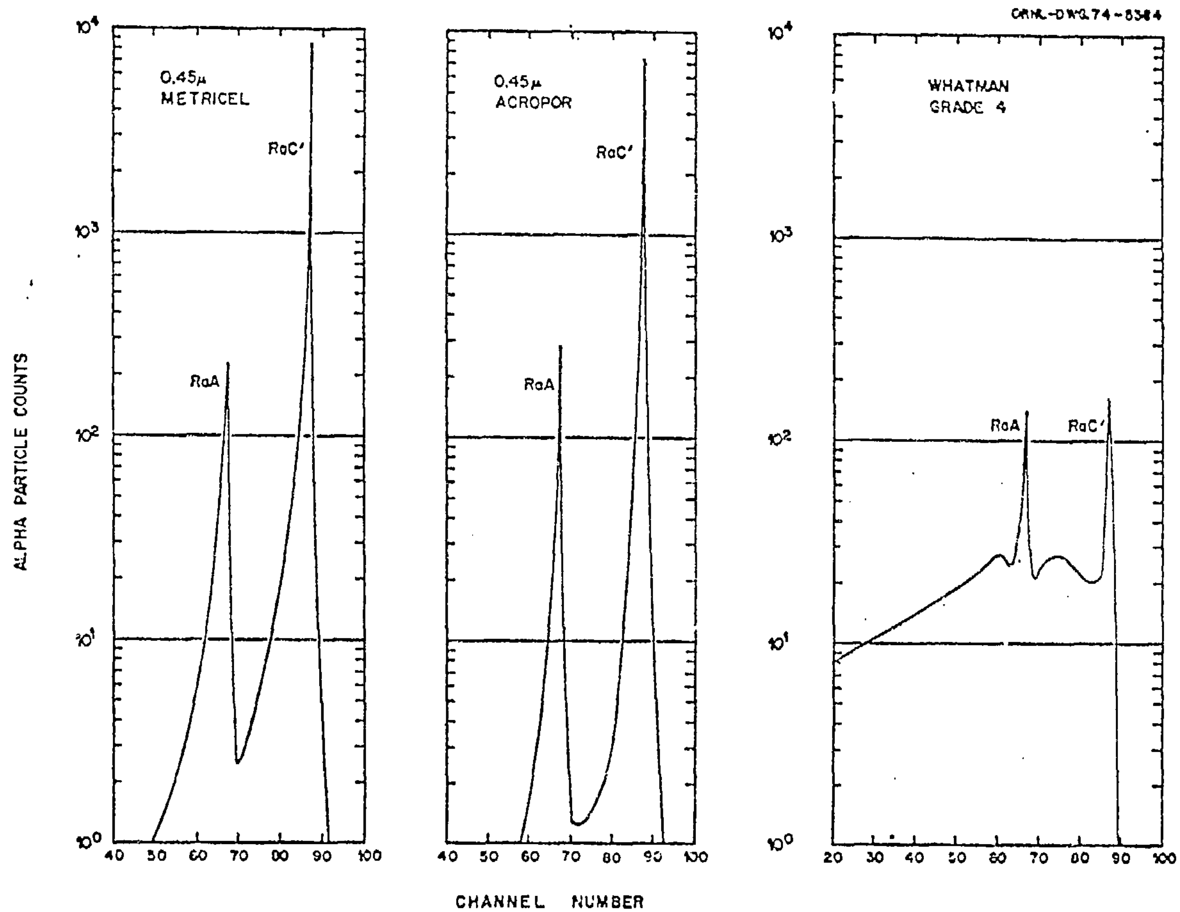
AIR
HELIUM
VACUUM
^{239}Pu
^{241}Am
^{233}U
ARBITRARY UNITS
10^0
10^1
10^2
10^3
10^4
200 200 300 320 340 360 360 400 420 360 400 420 440 460 340 360 380 400 420
CHANNEL NUMBER

Fig. 2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ALPHA PARTICLE COUNTS
0.45μ
METRICEL
RaC'
RaA
0.45μ
ACROPOR
RaC'
RaA
WHATMAN
GRADE 4
RaA
RaC'
CHANNEL NUMBER

Fig. 3

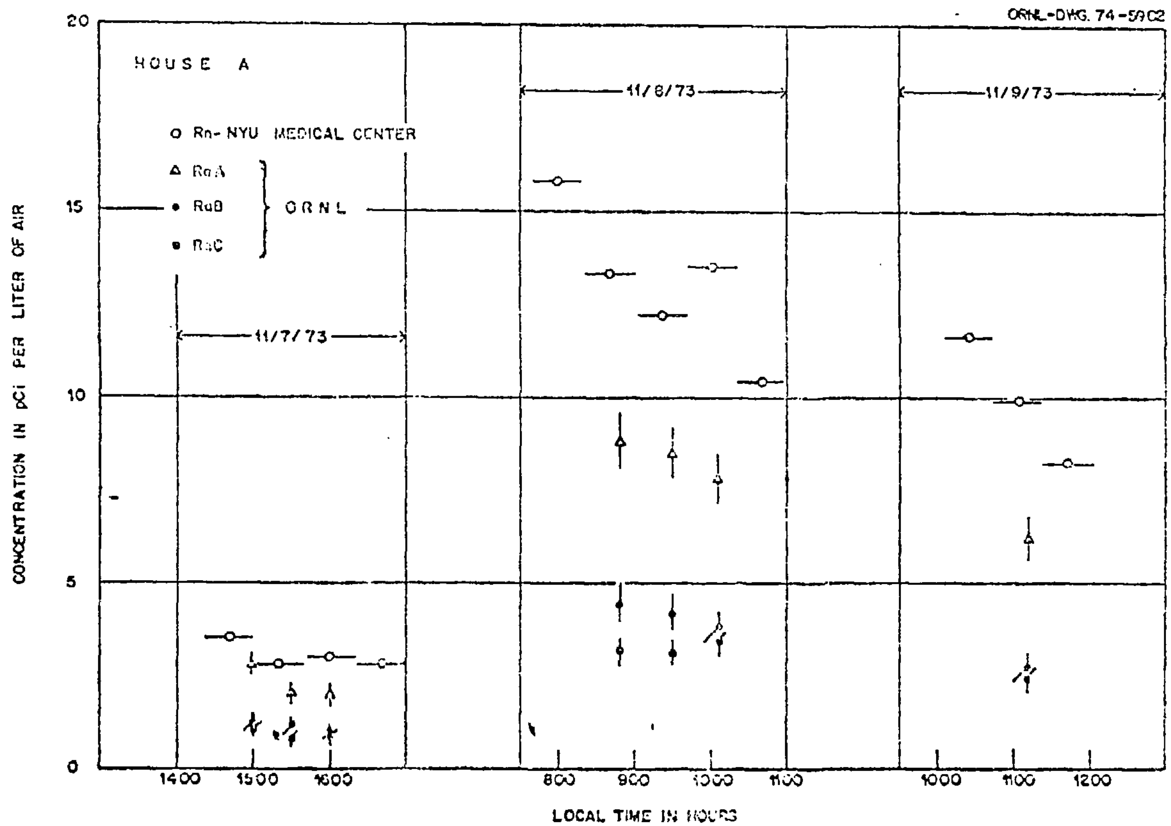
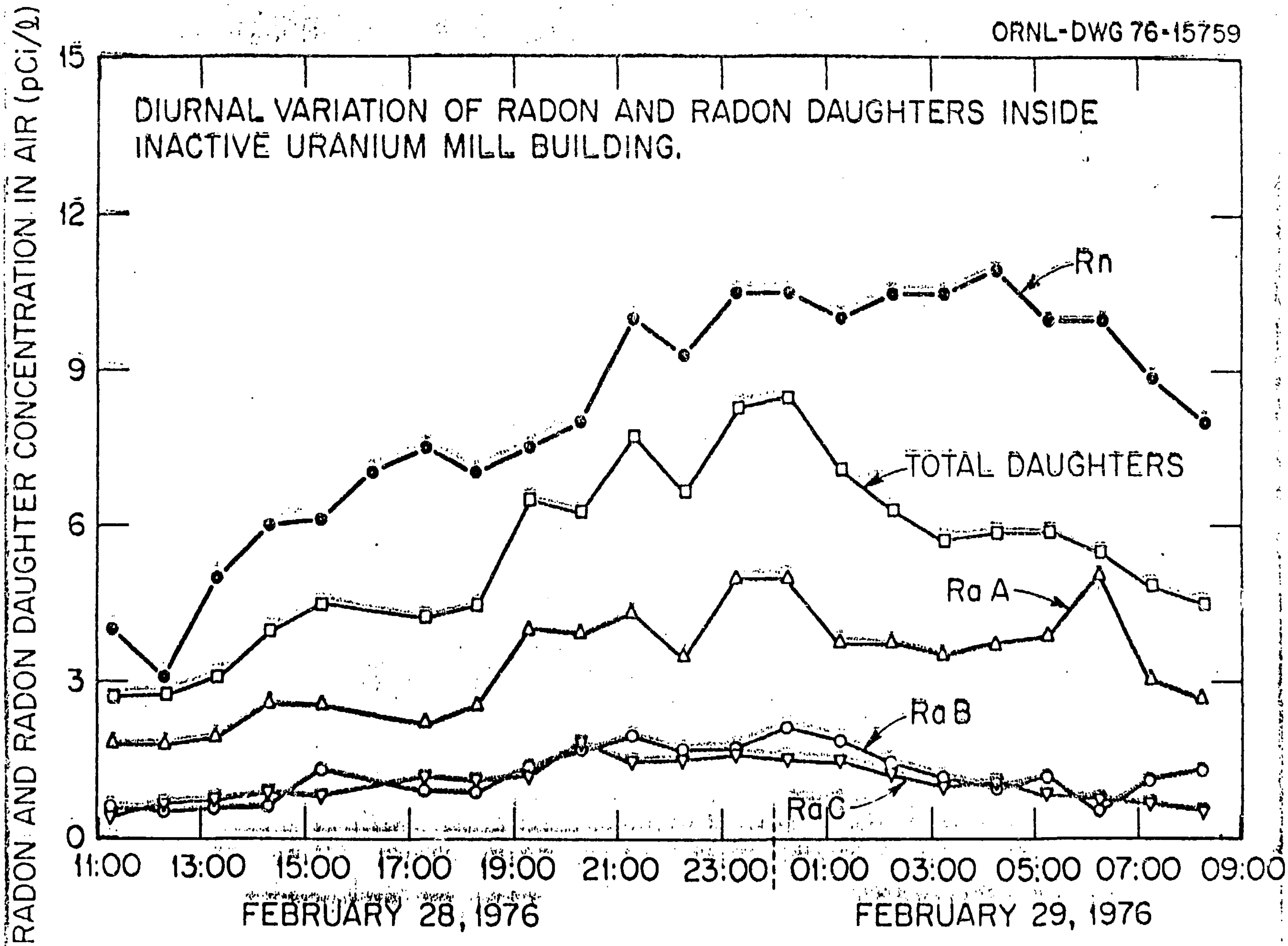

ORNL-DWG. 74-5902
HOUSE A
O Rn- NYU MEDICAL CENTER
Δ RaA
• RaB
• RaC
ORNL
11/7/73
11/8/73
11/9/73
20
15
10
5
0
CONCENTRATION IN pCi PER LITER OF AIR
1400
1500
1600
800
900
1000
1100
1000
1100
1200
LOCAL TIME IN HOURS

Fig. 4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DIURNAL VARIATION OF RADON AND RADON DAUGHTERS INSIDE INACTIVE URANIUM MILL BUILDING.
RADON AND RADON DAUGHTER CONCENTRATION IN AIR (pCi/ℓ)
15
12
9
6
3
0
Rn
TOTAL DAUGHTERS
Ra A
Ra B
Ra C
11:00
13:00
15:00
17:00
19:00
21:00
23:00
01:00
03:00
05:00
07:00
09:00
FEBRUARY 28, 1976
FEBRUARY 29, 1976

Fig. 5