Measurements of Radon-222 Concentrations in Dwellings in Great Britain

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ABSTRACT

The rate of emanation of ²²²Rn from room surfaces into room air was assessed in living rooms of 87 dwellings in England and Scotland. The mean rate of emanation of ²²²Rn found was 0.54 pCi liter⁻¹ hr⁻¹, but for rooms in which there was some exposed stone the mean was 2.5 pCi liter⁻¹ hr⁻¹. On the basis of a mean emanation rate of 0.54 pCi liter⁻¹ hr⁻¹, it is concluded that the current population exposure rate to the short-lived daughters of ²²²Rn for the population of Great Britain is 0.15 WLM/year. This exposure rate is dependent on the ventilation rates in dwellings and will increase as ventilation rates are reduced to conserve energy used for space heating.

In the sixteenth century Agricola (1556) wrote that miners in certain European communities were "... dying from the pestilential air they breath; sometimes their lungs rot away." Studies at the end of the nineteenth century and early twentieth century revealed that between 50 and 75% of deaths among working miners at Schneeberg, Germany, and Joachimstal, Czechoslovakia, was due to lung cancer (Harting and Hesse, 1879; Peller, 1939). Beginning in 1924 the noble gas ^{2 2 2} Rn was suspected of causing lung cancer in miners (Ludewig and Lorenser, 1924; Sikl, 1930; Saupe, 1933), but in 1951 Bale (unpublished memoranda quoted in Stewart and Simpson, 1963) demonstrated that 99% of the dose delivered to lung was due to the short-lived daughters of 222 Rn. These daughters are 218 Po (half-life, 3.05 min), ²¹⁴Pb (half-life, 26.8 min), ²¹⁴Bi (half-life, 19.7 min), and 214 Po (half-life, 1.6 x 10^{-4} sec). In view of the short half-life of ²¹⁴Po, it is invariably found in equilibrium with ²¹⁴Bi, and the alpha emission from ²¹⁴Po may be regarded as a prompt alpha

emission from ²¹⁴Bi. The hazards of exposure to the short-lived daughters of ²²²Rn were confirmed by an intensive study of the mortality of uranium miners in the United States (Lundin, Wagoner, and Archer, 1971) which revealed a strong correlation between the excess incidence of lung cancer and exposure to high concentrations of radon daughter products.

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1980

Uranium occurs widely in the earth's crust and consequently occurs in many materials used by the building industry. The fifth daughter product of ²³⁸U is ²²⁶Ra and this nuclide is also found in building materials and acts as a source for its decay product ²²²Rn, a gas which diffuses from soils and building materials into room air where it and its daughters are available for inhalation. The short-lived daughters attach to dust particles in the air and these, when deposited in the lung, may deliver a high dose to cells of the bronchial epithelium following the subsequent alpha decay of the attached ²¹⁸Po and ²¹⁴Po. Thus the population as a whole is also exposed to the short-lived daughters of radon. It is of interest to determine the current levels of this exposure in order to assess the implications of using waste materials that have a higher uranium concentration than traditional materials because of changes in building practice. One such material considered for use is gypsum, a by-product of the phosphate fertilizer industry. The effects of the use of by-product gypsum on the radon daughter concentrations in dwellings has been studied by O'Riordan et al. (1972), but at the time of this study no extensive survey of radon daughter concentrations in Great Britain had been made. To rectify this situation. Duggan and Bradford (1974) published the results of a preliminary survey of radon daughters in dwellings, and the work reported here is the result of an extension of the survey initiated by those authors.

METHOD OF MEASUREMENT

The radon concentration measured in both indoor and outdoor air is known to be subject to large temporal variation (Steinhausler, 1975). The concentrations of 222 Rn and its shortlived daughters within a room are predominantly determined by the rate of emanation of 222 Rn from room surfaces into the room air and the ventilation rate existing in the room. The emanation rate is mainly dependent on the radium content and porosity of the materials used in the construction of the building and of the ground beneath it. The effects of ventilation will be considered later. The measurements made during the survey reported here were designed to measure the rate at which 222 Rn was emanating from room

MEASUREMENTS OF RADON-222 CONCENTRATIONS

CLIFF

surfaces into room air in picocuries of ²²²Rn per liter of room air per hour. The direct measurement of radon gas concentrations at environmental levels is not easily accomplished with readily transportable equipment, and it was found easier to measure the ²¹⁸Po concentration. Measurements of ²¹⁸Po activity concentrations were made with the Radon Daughter Monitor (James and Strong, 1973) and the method of calculation described by Cliff (1978b).

It can be shown (Cliff, 1978a) that the rate of emanation of ²²² Rn can be found from

$$K = j\left(1 + \frac{j}{\lambda_A}\right) \left(C_A - C_A^{\dagger}\right) \quad pCi \text{ liter}^{-1} \text{ hr}^{-1}$$
(1)

where j = ventilation rate in room changes per hour, hr⁻¹

- $C_A = activity$ concentration of ²¹⁸ Po within the room, pCi liter⁻¹
- C_A^i = activity concentration of ²¹⁸ Po in the outside air, pCi liter⁻¹
- K = radon output of the room

and Eq. 1 is valid only if the air interchange occurs solely between the room and the outside air. To ensure that this was the case, rooms on the windward side of each dwelling were chosen for study. Equation 1 is also true only in the steady state, i.e., when the rate of the production of radon into the room is equal to the rate of loss resulting from radioactive decay and ventilation. The measurement of environmental concentrations of radon daughters is subject to statistical uncertainties that decrease as the concentrations increase. Thus, to reduce these uncertainties to a minimum, doors and windows were shut before measurements were made, and the concentrations of the nuclides within the room were allowed to increase. Some $2\frac{1}{2}$ hr must elapse between the reduction in ventilation and making the measurements if it is to be assumed that conditions have sensibly reached the steady state (Cliff, 1978a).

ORGANIZATION OF THE SURVEY

Since the main aim of this survey was to estimate the population exposure to the short-lived daughters of radon, the dwellings chosen were situated mainly in such areas of large population density as London, Leeds, Birmingham, Glasgow, and Edinburgh. In addition, some dwellings were measured at rural locations and some in areas where the 2^{2} Bu concentrations might be high because of the areas were Cornwall and Aberdeen. Where possible, dwellings were chosen which were typical of the housing stock for that area, although there was a higher proportion of dwellings of recent (post 1935) construction in the survey than in the total housing stock of Great Britain. Measurements were made in a total of 87 dwellings.

The main results of the measurements are summarized in the following sections. A more detailed description of the results has been published elsewhere (Cliff, 1978a).

RESULTS

The range of radon outputs measured was large (from 0.012 pCi liter⁻¹ hr⁻¹ to 5.5 pCi liter⁻¹ hr⁻¹), and the distribution was unsymmetrical. For this reason the distribution is shown on a logarithmic scale in Fig. 1. The median value of all the results is 0.32 pCi liter⁻¹ hr⁻¹ with a geometric standard deviation of 3.1. The mean for all the measurements is 0.60 pCi liter⁻¹ hr⁻¹, but the highest value recorded of 5.5 pCi liter⁻¹ hr⁻¹ (which is nearly twice the next highest value of 2.8 pCi liter⁻¹ hr⁻¹) occurred in a dwelling situated in an area of low population density, and this elwelling was constructed from local granite known to contain an elevated radium concentration. Such dwellings form a very low proportion of the housing stock of Great Britain, and for the purposes of estimating population exposure this value was elimi-



nated. The removal of this one high value reduces the mean value to 0.54 pCi liter⁻¹ hr⁻¹. When the dwellings surveyed are categorized by age or style (house, bungalow, or flat), there are no significant differences between the groups. As is to be expected, the factor most influencing the radon output of the room measured is the material used for the inner walls of the room. Those rooms having inner walls made of stone and plaster exhibit a higher radon output than those composed of other materials, such as brick and plaster, lathe and plaster, or plaster board. In particular, all rooms surveyed that had exposed stone within them had radon outputs which were elevated compared with rooms of similar construction without the exposed stone. The range of radon outputs found in rooms with some exposed stone surfaces is 0.80 pCi liter⁻¹ hr⁻¹ to 5.5 pCi liter⁻¹ hr⁻¹.

INTERPRETATION OF THE RESULTS

In a discussion of exposure to the short-lived daughters of radon, the usual unit of concentration for radon daughters is the working level (WL), which is defined as any combination of the short-lived daughters of radon (218 Po, 214 Pb, 214 Bi, and 214 Po) in 1 liter of air which will result in the ultimate emission of 1.3×10^5 MeV of alpha energy. Exposure is then measured in working level months (WLM) where the working month is taken as 170 hr. Thus exposure at 1 WL for 170 hr results in 1 WLM of exposure.

The mean radon output for dwellings in Great Britain is taken as $0.54 \text{ pCi liter}^{-1} \text{ hr}^{-1}$ on the basis of the survey results presented here, but the actual concentrations of the short-lived daughters of radon within a room with a given radon output is strongly dependent on the ventilation rate. Table 1 shows the activity concentrations of the short-lived daughters in a room with a radon output of 0.54 pCi liter⁻¹ hr⁻¹ when the incoming air has a radon concentration of 0.07 pCi/liter with activity ratios ²³²Rn : ²¹⁸Po : ²¹⁴Pb : ²¹⁴Bi = 1:1:0.8:0.6. These conditions for incoming air are typical for outside air as measured during this survey and agree with values quoted in the literature (Davies and Forward, 1970; Harley, 1973). Changes in the equilibrium conditions existing in the incoming air have little effect on the daughter concentrations within the room when the ventilation rate is below 1.5 room changes per hour. Ventilation rates measured in closed living rooms were often below 0.7 room changes per hour, and these are probably typical values during the winter heating season. Warren (1975) has reported the results of measurements of whole-house ventilation rates in six

TABLE 1

Radon Daughter Concentrations Within a Room Emanating 0.54 pCi liter⁻¹ hr⁻¹ for Various Ventilation Rates

Vantilation	Concentration, pCi/liter			Working
rate, hr	218Po	214 Pb	²¹⁴ Bi, ²¹⁴ Po	Po level, WL
0.1	5.07	4.76	4.55	0.0471
0.2	2.64	2.35	2.15	0.0230
0.5	1.10	0.85	0.69	0.0081
1.0	0.57	0.37	0.26	0.0035
1.5	0.39	0.23	0,15	0.0022
2.0	0.31	0.17	0.11	0.0016

dwellings and found the range to be 0.3 to 1.35 air changes per hour. If we assume a mean ventilation rate of 1 room change per hour then assume that people spend 80% of their time within buildings (occupancy factor 0.8, UNSCEAR, 1977), the mean population cumulative exposure rate is 0.144 WLM/year from exposure within buildings. The total mean cumulative exposure rate, including that received while in the open air, is then 0.15 WLM/year. The interpretation of this exposure rate must be approached with caution, Jacobi (1976) reviewed the data for the increased incidence of lung cancer in uranium and other miners and has derived a risk factor for this group of 200 excess lung cancers per 10⁶ persons per WLM. Reissland (1977) has applied this risk estimate to the population exposure derived from this survey and, on this basis, has predicted the incidence of lung cancer in women in Great Britain. As shown in Table 2, the predicted incidence exceeds the observed incidence for women of all ages below 40. This shows that the risk estimate derived from exposure of miners is unlikely to be applicable to the exposure of the population as a whole to radon daughters from building materials.

CONCLUSION

The mean population exposure to the short-lived daughters of ²²²Rn derived from this survey is 0.15 WLM/year, based on the assumption of a mean ventilation rate in dwellings of 1 room change per hour over the year. With the inevitable rise in energy costs and the consequent reduction in ventilation rates to reduce the costs of heating dwellings, this exposure will increase.

1265

CLIFF

TABLE 2

Predicted and Observed Incidence of Lung Cancer in Women in Great Britain*

-	Number of lung cancers per 100,000 persons				
Age		Predicted Incidencet			
years	Observed incidence	1 WLM/year	0.15 WLM/year		
20-24	0.1	2.4 .	0.36		
25-29	0.3	6.4	0.96		
30-34	1.0	10.4	1.6		
35-39	1.9	14.4	.2,2		
40-44	6.4	18.2	2.7		

*Data from J. A. Reissland, 1977.

†Based on 200 excess cancers per million persons per WLM.

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DISCUSSION

Ellett: You have shown the fallacy of applying absolute risk estimates based on male to a female population. However, Jacobi also calculated a relative risk (100% increase per 100 WLM) based on early miner data. Have you calculated the increased relative risk to women based on female lung cancer mortality data?

Cliff: I have not calculated the relative risk for women because this concept is population dependent and using the relative risk derived from a male population and applying it to a female population is just as fallacious as using absolute risk.

Jonassen: Your average radon output of 0.54 pCi liter⁻¹ hr⁻¹ will with an 0.5 volume/surface ratio correspond to an exhalation rate of 1300 atoms m⁻² s⁻¹, which is what we find for alum shale aerated concrete. Would you care to comment on that?

Cliff: If we consider that the only source of emanation is room surfaces, then I agree that $0.54 \text{ pCi liter}^{-1} \text{ hr}^{-1}$ is supported in a room of an $0.5 \text{ volume/surface ratio by an exhalation rate of 1300$ atoms m⁻² sec⁻¹. However, the model used to derive emanationrate does not assume this. All the model states is that within theroom there is a source or sources which together result in the $injection of <math>0.54 \text{ pCi of } ^{222}$ Rn per liter of room air per hour. Most of the rooms investigated were situated on the ground (first) floor where a major source would be ingress through cracks in the floor from the subsoil. However, since rooms with exposed stone surfaces yielded generally higher radon emanation rates it would appear that exposed stone is a large contributory factor. The fact that exposed suggests that further investigation of the sources of radon in room air is required.

Wrenn: Does your calculation of efflux rate (pCi/hr^{-1}) depend on the assumption that the only mechanism of removal of RaA is by dilution ventilation? If so, how might this affect your results?

Cliff: My calculation does assume that the removal of RaA and other daughters is purely by ventilation and radioactive decay. Since ventilation rate was recorded at the time that measurements were made of RaA, RaB, and RaC. I have looked at 18 of the results and calculated the equilibrium factor on the basis of the ventilation rate measured and compared it with the equilibrium 'factor actually measured from the daughter concentrations actually found. The mean of the ratios of these two results was 0.95 for the 18 cases studied. In five of the cases, the equilibrium factor derived from the daughter concentrations measured was lower than that predicted with the ventilation rate used as the sole means of daughter removal. This indicates that convective diffusion of daughters to walls may be a method of removal. However, the statistical uncertainty in the ratio derived from the two equilibrium factors calculated was usually about 50%. Thus this work is inconclusive, and I intend to carry out further studies on the effects of plateout of daughters onto walls in dwellings.

Wrenn: (Comment) Some years ago we showed that radon daughters in enclosed spaces (mine areas actually) could decrease by moving air (i.e., convective diffusion of aerosols and daughters to walls). This was very dependent on the concentration of condensation nuclei, and the effectiveness of removal increased dramatically with decreasing condensation nuclei concentration.

Jonassen: Both radon- and decay-product measurements showed pronounced annual variations, in general, with higher values during periods of increased atmospheric stability (e.g., inversion layers) and vice versa.

For the network of uninhabited control stations ventilation conditions were defined since, during the course of the investigation, all openings (doors, windows) were closed and indoor atmospheric temperature and humidity were monitored. At the other selected inhabited test sites, measurements were carried out several times at different seasons and different ventilation conditions to account for the living habits of the inhabitants.

Cliff: The rate of ventilation was monitored indirectly via continuous measurements of atmospheric temperature and relative humidity indoors as well as outdoors.

Participant (EPA): A quality factor of 10 was used for alpha-dose

1271