

# **The Incidence and Origin of Radon and Its Decay Products in Buildings**

K D Cliff, J C H Miles and K Brown



# **National Radiological Protection Board**

Chilton, Didcot, Oxon OX11 0RQ

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ABSTRACT

The largest contribution to population exposure from natural radiation arises from the inhalation of the decay products of radon in indoor air. In most instances the dominant source of radon within buildings is the subjacent ground. However, building materials do contribute to the radon concentration in indoor air, and in some circumstances may provide the major source. This report reviews some of the literature on radon emanation from soils and building materials world-wide. Details are given of measurements of radon emanation from common UK building materials, together with results of measurements of radon concentration in indoor air in sedimentary areas and in areas of enhanced uranium mineralisation. This work was part funded by a contract with the Building Research Establishment of the Department of the Environment (reference FS/3/399).

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## 1. INTRODUCTION AND MAIN CONCLUSIONS

This report details studies of the sources of the radioactive gases radon-222 and radon-220 found in air within all buildings. The main emphasis has been placed on radon-222 as this generally occurs in higher concentrations than radon-220, although the latter cannot be ignored. This work was part-funded by a contract with the Building Research Establishment, of the Department of the Environment (reference FS/3/399).

The principal isotope of radon, radon-222, has been known to exist in ambient air since 1901. Much higher concentrations of this gas and its decay products were found in uranium and some other mines. Workers in these mines were found to experience an excess incidence of lung cancer and the causative agent of this disease is considered to be the decay products of radon-222, namely polonium-218, lead-214, bismuth-214 and polonium-214, which are referred to as the short-lived decay products of radon-222 since none has a half-life exceeding 30 minutes. The two polonium isotopes decay by emitting an alpha particle and it is these particles that deliver a relatively high radiation dose to small volumes of tissue. Polonium-214 is formed by the decay of bismuth-214, but has a half-life of only 164  $\mu$ s. In all practical situations polonium-214 is in equilibrium with bismuth-214 and bismuth-214 acts as though it decays by beta and gamma-decay followed by a prompt alpha-particle emission. Thus, the short-lived decay products of radon-222 are often stated to be polonium-218, lead-214 and bismuth-214, the presence of polonium-214 in equal activity concentration to bismuth-214 being implied.

In 1956 a study of natural radiation in Swedish housing was published which noted the elevated concentration of radon-222 and its decay products in indoor air compared with those concentrations measured in the open air. Since then studies have taken place of the incidence of radon-222 and its decay products in dwellings and other buildings in a number of countries including the UK. In most countries the vast majority of buildings sustain radon-222 concentrations between a factor of two and ten higher than the concentrations in the open air.

In a number of countries situations were discovered that resulted in unacceptable radon-222 decay products exposure within dwellings and other buildings. These include the misuse of mining waste (the use of mill tailings in construction in Colorado), building on reclaimed land of enhanced radium-226 concentration in Florida, building in areas where the geology includes widespread uranium mineralisation, as occurred in the uranium mining communities of Canada, and the use of a traditional raw material for construction but in a novel form, such as light-weight alum-shale concrete in Sweden. In USA, Canada and Sweden remedial action has been undertaken in limited circumstances to reduce the concentrations of radon-222 and its decay products within buildings to below a prescribed level.

The first studies of radon-222 and its decay products in buildings assumed that the major source of radon was the trace quantity of radium-226 in most traditional building materials. It was soon realised, however, that the concentrations of these nuclides measured in room air could not be supported solely by the radium-226 content of the building elements. Further work demonstrated that for single family dwellings (houses, ground-floor flats and bungalows) the dominant source of radon-222 was the ground beneath the structure. However, the contribution from the building materials, although comparatively small, cannot be ignored. The relative strengths of the various sources of radon-222 in dwellings depends upon the form of construction, the building materials used and the local geology.

The largest survey of radon-222 decay products concentrations in UK dwellings was carried out in 1976, but measurements were made in only 80 dwellings out of the UK housing stock of about 20 million. This survey, therefore, concentrated on the major centres of population in order to determine a representative figure for the mean population exposure to the decay products of radon-222. However, some measurements were also made in rural areas. A major difference between other forms of exposure to natural radiation and that due to radon-222 decay products was immediately apparent from the survey. For cosmic-ray exposure the variation in the UK is within  $\pm 10\%$  of the mean, for terrestrial gamma-ray exposure the range is probably within a factor of three either side of the mean, but for radon-222 decay product exposure the range is probably within two orders of magnitude either side of the mean.

The wide range of potential exposures to the decay products of radon-222 and the limited data available on the relative strengths of common building materials as sources of radon-222 led to the institution of the studies reported here. Apart from studies on building materials in the laboratory, measurements were made of the concentration of radon-222 decay products in various offices of a large complex offering a diversity of office size, construction materials and elevation. The concentrations of radon-222 and its decay products in indoor air in regions where the local geology suggested that the concentrations of these nuclides in air might be much higher than the UK average were also measured. Details of the entire measurement programme are given in the main text of the report (Sections 2-6) together with a review of data reported by other workers where this is relevant.

The radon-222 emanation rates from 165 specimens of building material were determined. The specimens ranged through relatively common materials, such as Fyfe-stone blocks, insulating blocks, bricks (silica and clay), light aerated concrete, etc, but included some items of less usual composition such as brick composed of oil shale. In the text the radon-222 emanation rates are given in terms of per unit area, per unit volume and per unit mass. The range in emanation rates per unit area for the materials reported here is  $3.6 \mu\text{Bq m}^{-2} \text{ s}^{-1}$



(for a clay brick) to  $790 \mu\text{Bq m}^{-2} \text{ s}^{-1}$  (for a Fyfe-stone block). Fyfe-stone block is seldom used to line room surfaces, but even this material has radon-222 emanation rates which are much lower than those of light-weight concrete containing alum-shale of the type manufactured in Sweden prior to 1975. This material is reported to have an emanation rate of  $8600 \mu\text{Bq m}^{-2} \text{ s}^{-1}$  and it is unlikely that any current or proposed building material would approach this figure. To illustrate the worst-case contribution of building materials to radon-222 in room air a room constructed entirely from this light-weight concrete containing alum-shale can be considered. Taking a room with a surface to volume ratio of  $1.8 \text{ m}^{-1}$  this would result in a radon-222 concentration in the room of  $56 \text{ Bq m}^{-3}$  at a ventilation rate of one air change per hour. This concentration is frequently exceeded in UK dwellings of quite normal construction. This does not, of course, indicate that the building elements play no, or little, part in contributing to the radon-222 concentration in indoor air. In some areas, where the local subsoil contains little radium-226, the building materials may be the only significant source of radon-222, but here the concentration of radon-222 in room air will be low and of no consequence as a potential health hazard. This illustration does, however, emphasise that in the vast majority of cases the ground subjacent to the building is the major source of radon-222 found within the building.

One brick, having oil shale as a main constituent, had a measured radon-222 emanation rate of  $290 \mu\text{Bq m}^{-2} \text{ s}^{-1}$ . This exceeded, by a factor in excess of two, the highest emanation rate of any brick of more traditional manufacture. Other materials that have an enhanced radium-226 concentration, such as phosphogypsum from the phosphate fertilizer industry, have been suggested for use in buildings in the UK. To date there has been minimal market penetration for this product although it is widely used in continental Europe. There are other materials, such as red mud from bauxite processing, which would be suitable in many respects for use as building materials but for their relatively high radium-226 concentration. In all judgements concerning the radium-226 concentration of building materials it must be remembered that the radium-226 concentration alone does not indicate the strength of that material as a source of radon-222. Another parameter required is the fraction of the radon-222 formed within the material that escapes from the surface. If the fraction of radon-222 escaping is low this will result in a higher gamma-dose rate from the material than from a material of similar radium-226 concentration but of higher emanating fraction. Emanating fractions in the range 0.3–12% were measured during the work reported here, with the values for the most commonly used materials being less than 4%.

Measurement programmes involving active air sampling equipment and using passive radon detectors have shown that the distribution of radon-222 decay products concentrations in room air of dwellings in the relatively few areas of enhanced uranium mineralization is quite different from that found within

dwellings situated in the major centres of population where the geology is predominantly sedimentary. In rare cases, the annual effective dose equivalent may exceed 100 mSv. The exposures can be reduced, but not without financial costs. The simplest action to reduce the concentration is to increase the ventilation rate but at the expense of increased space-heating costs. Alternatively, electrostatic precipitators can be installed as these have been shown to be effective in reducing the concentration of the decay products of radon-222 in room air (see Figure 8, p58). However, although the running costs of electrostatic precipitators are low the expense of their purchase is quite high. In these areas, it is the soil beneath and around the building that has enhanced radium-226 concentrations and is the source of the high concentrations of indoor radon-222. Studies are in hand to determine ways of preventing the ingress of radon into houses. It should be noted that as far as new buildings are concerned work carried out in the USA and Canada has shown that on soil of any uranium concentration it is possible to construct a 'radon free' building without departing too far from traditional building practice.

Measurement of radon-222 decay products concentrations and ventilation rate under conditions where all exterior doors and windows are closed but all internal doors open has been shown to produce the correct estimate of the whole house radon-222 production rate. However, such a measurement does not permit a realistic estimate of the likely radon-222 concentration to be found in any particular room under normal living conditions. On the other hand, when measurements are made in a single closed room there is a systematic error in measuring the air exchange rate as the degree of air exchange between adjacent rooms is difficult to control and is not simply quantified. Nevertheless this latter method of measurement will, under most circumstances, allow a reasonable prediction of the radon-222 concentration in the room under varying ventilation conditions. Thus single-room studies are to be preferred when using short-period active surveys to determine the likely long-term average radon-222 concentration in dwellings.

It being generally recognised that the dominant source of radon-222 in buildings is the ground subjacent to the building, suggestions have been made that radon-222 concentrations should be lower on floors of higher elevation than those on or near the ground. Measurements in high-rise buildings, however, have shown that this is not generally the case. Common services are often taken from floor to floor through ducting, and stairwells and lift shafts link different floors providing easy routes by which radon-222 can pass from lower to higher floors. Thus it is unlikely that occupants of flats on the upper floors of high rise buildings receive a significantly lower exposure to radon-222 decay products than do the occupants of flats closer to the ground.

For many people employed in offices a significant proportion of their exposure to the decay products of radon-222 could occur at work. Although the

data presented in this report covers only a limited number of offices, these offices were diverse in size, elevation and construction materials. The radon-222 decay products concentration was not related systematically to floor level and confirms the statement in the last paragraph. Excepting two offices with air conditioning using a high percentage of make-up air the remainder exhibit quite a wide range of radon-222 decay products concentrations, but the mean of the recorded values corresponds to just under 80% of the mean value found in ground-floor living rooms of houses built on the same soil type as the office block. This data might suggest that office workers have a reduced exposure to the decay products of radon-222 while at work than they do at home. However, the difference, if real, is small and tentative in view of the small sample sizes of both dwellings and offices. There would seem no firm evidence to suggest that radon-222 decay products exposure rates at work for office workers are significantly different from those at home.

Theoretical considerations have led to the postulation that under certain circumstances, particularly at high ventilation rates, the working level (WL) concentration in room air due to the decay products of radon-220 should exceed that due to the decay products of radon-222. Measurements carried out during this programme of work have not borne this suggestion out, although the decay products of radon-220 are present in room air and the concentrations found suggest that the exposure of the public to these decay products is non-trivial. More data on the incidence of the decay products of radon-220 will be collected over the next few years as part of the Board's continuing programme of evaluating the exposure of the general population to all sources of natural radiation.

A more detailed exposition of the above is given in the remainder of the report which is organised into the following sections.

Section 2 presents some background information, discusses the other naturally occurring isotopes of radon and introduces the special units used to describe the concentration in air of radon decay products and the unit commonly used to measure exposure to these nuclides.

Section 3 deals with radioactivity in building materials and reports values measured in other countries as well as those found during the course of the studies covered by this report.

Section 4 covers the exhalation of radon from soils and rocks and common building materials together with the effect of surface coatings in limiting the ingress of radon into room air.

Section 5 presents measured values of radon-222 and radon-220 decay products in air from several countries but deals in some depth with the values found in this study.

Section 6 reviews the theoretical aspects of radon evolution into the pore spaces of materials and of the transport of radon through materials.

A number of appendices are attached for the benefit of readers who are

likely to come from a broad spectrum of disciplines, some of whom may not be familiar with the terminology of radiological protection nor with the common measurement techniques in this field.

Appendix A describes in detail the decay chains in which the natural isotopes of radon occur and gives values for the energies and types of emission of the decay products of radon-220 and radon-222. An expanded exposition of the origin of the working level (WL) and working level month (WLM) units is given which may be easier to follow than the cursory treatment in Section 2. Factors for converting from the units of exposure peculiar to radon to the more familiar units of dose-equivalent are also given, thus permitting risks from exposure to radon to be compared directly with the risks from exposure to other forms of ionizing radiation.

Appendix B presents details of the measurement techniques used during this programme of work and the interpretation of results.

Appendix C expands the information presented in Sections 3 and 4 on the radioactivity in building materials and on radon emanation from these materials. In this appendix the chemical constitution and method of manufacture of those materials for which the manufacturers have supplied the information are given.

## 2. BACKGROUND

Five years after Becquerel's discovery of radioactivity in 1896, measurement of radioactivity in ambient air was reported by Elster and Geitel<sup>(1)</sup>. Thus commenced studies of atmospheric radioactivity which have continued with increasing intensity to the present. Close to the ground and within buildings the predominant sources of airborne radioactivity are isotopes of the noble gas radon and their decay products. A detailed discussion of the naturally occurring isotopes of radon together with an explanation of the special units adopted for the measurement of the concentration of the airborne decay products of radon are presented in Appendix A. The three naturally occurring isotopes of radon are radon-219, radon-220 and radon-222, known historically as actinon, thoron and radon, respectively.

This report is predominantly concerned with sources and levels of radon-222 in the atmosphere, but some discussion of radon-220 and its decay products in the general environment is also included. Apart from a few special environments the concentration of radon-219 in ambient air is very low and contributes little if any to human exposure (see Appendix A). Hence radon-219 is not considered in the body of this report.

The primordial nuclides heading the decay chains in which radon-222 and radon-220 occur are uranium-238 and thorium-232, respectively. These nuclides occur widely in nature, and all soils and rocks contain them in varying concentrations. Most traditional building materials are derived from rocks and soils and hence they too contain uranium and thorium, albeit usually in trace quantities. The immediate precursors of radon-220 and radon-222 are radium-224

and radium-226, respectively. In most cases these isotopes of radium are in radioactive equilibrium with the thorium-232 and uranium-238 in the raw material used to fabricate building materials such as bricks. In most cases there is little chemical separation of these species as the materials are processed. The equilibrium condition then persists in the final products.

Soils, rocks and building materials are porous, and when a radium atom close to the surface of a pore space decays to form the gaseous element radon, this can diffuse into the pore space and ultimately through interlinked pore spaces to the surface and into the surrounding air. There is, however, a major difference in half-life between radon-220 and radon-222. Radon-220 has a half-life of only 54.5 s, whereas that of radon-222 is 3.82 days. Radon-220 released to the pore space at an appreciable depth within the material will decay before reaching the surface and will not contribute to the airborne concentration of radon-220. Radon-222, on the other hand, can migrate considerable distances before decaying. The release of radon-220 to the environment is therefore a surface-layer effect, whereas the release of radon-222 is a bulk effect. In most environments the activity concentration of radon-222 exceeds that of radon-220, but the decay products of the latter do contribute to human exposure.

Radon released from the ground is rapidly diluted by the atmosphere due to turbulent mixing and in most cases the concentrations in outside air of radon-222 and its decay products are low, being of the order of  $2.6 \text{ Bq m}^{-3}$  in the UK. Within buildings the radon mixes with the volume of air contained by the building, and is further diluted with outside air exchanging, by ventilation, with the internal air. Under these conditions the concentration of radon-222 in indoor air is usually higher than that in outside air even at quite high ventilation rates of two to three air changes per hour. When ventilation rates are reduced to well below one air change per hour the internal concentration of radon-222 and its decay products often exceed the concentration of radon-222 in outside air by a factor greater than ten.

Miners in some areas of Europe have been reported as dying of lung disease since the 16th century. Evidence obtained at the end of the last century and during the early years of this century strongly suggests that this disease was lung cancer<sup>(2,3)</sup>. A large excess incidence of lung cancer was noticed among uranium miners in USA and Czechoslovakia in more recent times, and extensive epidemiological studies have been carried out on these groups<sup>(4-11)</sup>. At first, the causative agent of these lung cancers was thought to be the elevated concentrations of radon-222 known to exist in mine air. However, in 1951 it was shown that almost all the dose to lung tissue resulting from the inhalation of radon-222 laden air was delivered by the decay products of radon-222 rather than the radon-222 itself<sup>(12)</sup>. Because of their short half-lives these decay products rapidly appear in a stream of fresh radon such as that released from an ore body. In all practical situations radon-222 laden air also contains the short-lived

decay products, and the activity concentrations of the decay products may approach that of the parent radon in air if ventilation rates are low. The short-lived decay products of radon-222 are heavy metals which, when first formed in air, exist as charged ions that rapidly neutralize and attach to sub-micron particles in the air. When inhaled these decay products are deposited on the walls of the airways in the lungs and may decay there. It is the alpha particles that deliver high dose equivalents to small volumes of tissue and are therefore likely to be the cause of the excess lung cancers in those exposed to the short-lived decay products of radon-222. A special unit, the working level (WL), was introduced as a measure of the potential alpha-energy concentration in air (see Appendix A). The working level is defined as any concentration of the short-lived decay products of radon-222 (polonium-218, lead-214, bismuth-214 and polonium-214) in one litre of air that will result in the ultimate emission of  $1.3 \times 10^5$  MeV of alpha-particle energy. A unit of exposure was then devised based on a working month of 170 hours duration. The unit of exposure is the working level month (WLM) and an exposure to 1 WL for 170 hours gives an exposure of 1 WLM. Likewise an exposure to 2 WL for 85 hours also results in an exposure of 1 WLM and so on.

In the epidemiological studies on uranium (and other) miners the cumulative exposures have been given in terms of WLMs and this has made comparison with other radiation exposures, where doses are usually expressed in grays (Gy) or dose equivalents in sieverts (Sv), difficult. The International Commission on Radiological Protection (ICRP), in its latest recommendations, introduced the concept of effective dose equivalent to be used when the body is non-uniformly irradiated<sup>(13)</sup>. The effective dose equivalent (Sv) is that dose equivalent which if delivered uniformly to the whole body would result in the same total detriment to the individual as results from the actual dose equivalent delivered to the organ in question (here the lung). By examining the epidemiological data and using realistic lung model techniques to calculate the dose to the various regions of the lung, ICRP produced a report on the limits for inhalation of radon daughters by workers. In this report they conclude that an exposure of 1 WLM to the short-lived decay products of radon-222 for workers results in an effective dose equivalent of 10 mSv<sup>(14)</sup>.

As mentioned earlier radon-222 and its decay products usually occur at low concentrations in the open air and at more elevated concentrations within buildings. Where the induction of cancer has been shown to be the consequence of exposure to ionizing radiation at high doses or dose rates it is assumed that this effect will also occur at low doses or dose rates, but with reduced frequency. This is held to apply down to zero dose. It would seem consistent, therefore, to assume that exposure of the population to the short-lived decay products of radon-222 in the general environment might be responsible for some of the lung cancer incidence in the general population. Lung cancer induction being

the deleterious effect known to occur in miners exposed to much higher concentrations of radon-222 decay products. However, extrapolation from the data for miners to the situation of exposure within buildings is less straightforward than similar exercises in extrapolating from effects at high dose to those at low dose for exposures to gamma-rays or X-rays. One problem is that the miner population is all male, and another is that they are simultaneously exposed to other agents in mine air which may act as co-carcinogens. A major difference, however, is in the level of physical activity undertaken by miners compared with the more sedentary office occupations and pastimes in homes. Miners have a higher average breathing rate than most other persons exposed to radon-222 decay products. The dose to lung tissue resulting from the inhalation of radon-222 decay products decreases as the breathing rate is reduced but not on a pro-rata basis. Considerations of differences in breathing rate, together with the known incidence of lung cancer in the general population prior to the large increase in incidence attributable to tobacco smoking, have resulted in the suggestion that the upper bound for the effective dose equivalent resulting from an exposure of 1 WLM in the general environment is 5 mSv<sup>(15)</sup>. This figure is used within this report in order that a comparison of exposures from other natural radiation sources can be made.

No epidemiological data exists concerning the health effects of exposure to the decay products of radon-220, but as these decay products also result in alpha irradiation of lung tissue their effect should be similar to those of the decay products of radon-222. However, there are significant differences in the properties of the two radon series which affect their respective dosimetry.

The half-life of 3.82 days for radon-222 is long compared with its first four decay products, and these decay products in air are unlikely to be very far removed from equilibrium with the parent gas. Under these circumstances it can be demonstrated that nearly all the dose to lung tissue arises from the inhaled decay products. In the case of radon-220, with a half-life of 54.5 s, this will be close to equilibrium with its decay product, polonium-216 of half-life 0.16 s. However, the next member of this decay scheme is lead-212 with a half-life of 10.6 h and, under all practical ventilation rates, lead-212 and bismuth-212 will be far from equilibrium with the parent radon-220. This indicates that in some situations the dose to lung from airborne radon-220 may be significant. In the general environment the concentration of radon-220 and polonium-216 are low and the doses resulting from inhaling these nuclides is negligible. In industrial circumstances where large concentration gradients of radon-220 may exist close to thorium bearing materials the dose from inhaled radon-220 and polonium-216 may be of similar magnitude to that of the later decay products lead-212 and bismuth-212<sup>(16)</sup>. In the general environment it is the inhalation of the decay products of radon-220 (lead-212 and bismuth-212) which pose a potential hazard, and here the second difference between the decay products of radon-222 and radon-220

manifests itself. The longest half-life of the short-lived decay products of radon-222 is 26.8 minutes (lead-214) and hence the decay products of radon-222 deposited in the bronchial tree will largely decay in lung before biological removal mechanisms are effective. However, lead-212 has a half-life of 10.6 h and that of bismuth-212 is 60.5 minutes. When these nuclides are deposited in lung a substantial proportion of the deposit will be cleared from lung by biological mechanisms. These include translocation to blood and removal via the mucus sheath that is moved towards the throat by the ciliated epithelium of the upper airways of the lung. In this way dust and other particles are carried to the throat where they are swallowed and passed to the gut. Thus, in the case of inhaling air containing the decay products of radon-220 having a potential alpha-particle energy concentration of 1 WL, the dose to lung tissue will be less than that for 1 WL, from radon-222 decay products, but a non-trivial dose may be delivered to organs other than lung<sup>(17)</sup>.

For workers, ICRP have assigned a risk per unit WLM exposure to the decay products of radon-220 that is one third of the risk of an equivalent exposure to the short-lived decay products of radon-222<sup>(14)</sup>. No similar assessment has been published for environmental exposures to the decay products of radon-220, however, there are no sound reasons to justify changing the factor of three between the risks from radon-220 decay products exposure and those from radon-222 decay products. On the other hand, the uncertainties in the derivation of dose conversion factors applied to radon decay products should not be understated and it would be misleading to state WLM to effective dose equivalent conversion factors to more than one significant figure. It is reasonable, therefore, to take a value of 2 mSv as the upper bound for the effective dose equivalent resulting from an exposure of 1 WLM to the decay products of radon-220.

### 3. RADIOACTIVITY OF BUILDING MATERIALS

#### 3.1 Early values reported in the literature

There has been little published work concerning the radionuclide content of building materials originating from UK raw materials. The radionuclides of general interest are potassium-40, radium-226 and thorium-232 as these (or their decay products) contribute to the terrestrial gamma radiation to which all populations are exposed. Natural potassium contains 0.012% of the radioactive isotope potassium-40. As potassium is an essential requirement in diet, potassium-40 acts as an internal as well as an external radiation source. Radium-226 and thorium-232 are the sources of radon-222 and radon-220, respectively, and are of greater interest in the context of this report. However, where authors have given values for the three nuclides these have been presented below for information.

Table 1 displays reported values for building materials actually used in the UK and for some materials that may be used in the future.



Table 1  
Radionuclide concentrations in some UK building materials

Material	Mean specific activity (Bq kg <sup>-1</sup> )			Number of specimens	Reference
	Potassium-40	Radium-226	Thorium-232		
Clay bricks	620	52	44	23	(18)
Calcium silicate bricks (rock aggregate), china clay	790	52	3.9	3	(18)
Calcium silicate bricks (gravel aggregate)	350	7.4	3.4	5	(18)
Calcium silicate bricks (flint aggregate)	12	2.2	5.4	5	(18)
Granite 'bricks' (reformite)	590	11	13	3	(18)
Granite 'bricks'	1000	89	81	7	(18)
Phosphate ore (source material for type A gypsum)	260	1400	51	14	(18)
Gypsum type A (Waste product super-phosphate fertilizer)	62	790	17	6	(18)
Gypsum type A from carbonatite ores	nd	120	23	1	(18)
Gypsum type B (natural UK gypsum)	140	23	7.5	69	(18)
Cement from type B gypsum	0	7.4	0	1	(18)
	0	210	55	1	(18)
Concrete block/fly ash (type 1)	560	7.4	34	1	(18)
Concrete block/fly ash (type 2)	530	140	33	1	(18)
Concrete block/fly ash (type 3)	210	52	44	1	(18)
Aerated concrete	650	89	13	2	(18)
Vermiculite	1400	93	0	1	(18)
Sand and gravel	33	4	7	10	(19)
Cement	160	22	18	6	(19)
Phosphogypsum from sedimentary ores	41	630	18	60	(19)
Weald clay brick with 10% high carbon grit	nd	39	nd	1	(20)
London clay brick containing 50% by volume PFA	nd	78	nd	1	(20)
Gault clay brick with 5-6% by volume of high carbon grit	nd	23	nd	1	(20)

nd = not determined.

In Table 1 many of the sample sizes are too small to permit general conclusions to be drawn. In most cases the variability between specimens of the same material are large (highlighted by the two specimens of cement from type B gypsum<sup>(18)</sup>) and for this reason the mean specific activities have been given to two significant figures only.

Values of the specific activities of building materials commonly used in other countries have been reported and are shown in Table 2<sup>(21)</sup>.

The differences in specific activity of similar materials from different points of origin, shown in Tables 1 and 2, are not marked when the standard deviation between specimens is considered. For the clay bricks reported in Table 1 for the UK, the standard deviation on the specific radium-226 activity given in the reference is  $\pm 26 \text{ Bq kg}^{-1}$ <sup>(18)</sup>, admittedly for a relatively small sample size. The mean range of reported specific radium-226 activity is 23–100  $\text{Bq kg}^{-1}$  from sample sizes of one and six, respectively. If the relative standard deviation on the reported mean specific radium-226 activity is similar to that above ( $\pm 50\%$ ) then clay bricks, regardless of point of origin, have a probable mean specific radium-226 activity of  $63 \pm 31 \text{ Bq kg}^{-1}$ .

Tables 1 and 2 reveal certain materials with enhanced radium-226 activity, and although many are restricted to locations where they occur naturally others arising as by-products of certain extensive industries could find world-wide application. Two examples of the latter materials are phosphogypsum, which occurs as a by-product in the manufacture of phosphate fertilizer, and red mud arising from the production of alumina from bauxite. From Tables 1 and 2 it can be seen that phosphogypsum typically contains about 600  $\text{Bq kg}^{-1}$  of radium-226 (excluding the very low value reported from Sweden which suggests that the phosphogypsum has been mixed with natural gypsum). This is an order of magnitude higher than clay bricks and although the structural and aesthetic properties of phosphogypsum are indistinguishable from those of natural gypsum its wide application in the building industry should not be encouraged. For materials of similar density and porosity the rate of emanation of radon-222 will be proportional to the radium-226 specific activity. Phosphogypsum has been widely used in continental Europe but penetration of the UK market has been minimal. The authors are not aware of any use of red mud bricks in the UK, but fly ash of similar specific radium-226 activity is widely used as an aggregate in this country.

In Table 2 the results are given of measurements on some materials that have produced problems of enhanced radon-222 concentrations in certain areas or countries. Alum-shale of enhanced radium-226 concentration occurs widely in Sweden and has been used as a building material for centuries. With the advent of foaming agents permitting the manufacture of light-weight aerated concrete containing alum-shale, a radiological problem was created. The aerated concrete is much more porous than normal concrete. This allows a much larger fraction of

**Table 2**  
**Radionuclide concentration in building materials used outside the UK**

Country	Material	Mean specific activity (Bq kg <sup>-1</sup> )			Number of specimens
		Potassium-40	Radium-226	Thorium-232	
Finland	Building sand and gravel	1030	37	43	166
	Clay bricks	960	78	62	33
	White bricks	580	22	23	3
	Cement	240	44	26	7
	Aerated concrete	360	49	36	2
	Natural gypsum	25	7	2	1
	Slag aggregate	190	100	69	3
Federal Republic of Germany	Building sand and gravel	240	< 15	< 18	50
	Granite	1300	100	80	32
	Bricks (traditional)	670	59	67	109
	Pumice-aggregate concrete blocks	770	74	80	31
	Slag-aggregate concrete blocks	530	150	100	9
	Portland cement	240	< 26	< 18	14
	Natural Gypsum	96	< 18	< 10	23
	Phosphogypsum	96	590	< 16	33
	Red mud bricks	340	280	230	23
	Fly ash	720	210	130	28
Italy	Lithoid tuff	1500	130	120	-
	Nenfro (a variety of tuff)	2100	240	220	-
Norway	Brick	1100	100	62	6
	Concrete	720	26	36	4
	Light-weight concrete	240	33	26	5
	Clinker	820	96	60	6
	Natural gypsum	11	11	3	2
	Cement	240	30	18	4
Sweden	Concrete ballast	820	48	73	296
	Brick	960	96	130	-
	Cement	240	55	47	-
	Aerated concrete without alum-shale	1300	55	18	-
	Aerated concrete with alum-shale produced 1947-1975	770	1300	67	-
	Aerated concrete with alum-shale produced since 1974	530	330	28	-
	Gypsum plasterboard	22	4	< 1	-
	Phosphogypsum	48	15	62	-
	Light-weight aggregate	1000	140	160	-
USA	Phosphate land fill, Florida	-	740	-	-
	Gypsum from Florida phosphate rock	-	1200	10	-
	Uranium mill tailing	-	600	-	-

the radon-222 formed within the aerated concrete to escape to the atmosphere, thereby increasing the concentration of radon-222 in indoor air. This problem was so acute that the use of light-weight aerated alum-shale concrete of the form manufactured since 1947 was banned as a building material in Sweden in 1974<sup>(22)</sup>.

Uranium mill tailings (the waste material from the processing of uranium ore) was used in Colorado as a land fill and back-fill material around buildings, particularly in Grand Junction and Durango. This resulted in the indoor concentrations of radon-222 and its decay products in houses built over mill tailings, or of construction incorporating mill tailings, being considerably elevated compared to non-affected housing in the area. The practice of using uranium mill tailings for these purposes was stopped in 1966 and a programme of remedial action undertaken<sup>(23)</sup>.

Extensive phosphate ore reserves occur in Florida and the dominant mode of extraction is by strip-mining. This involves removing the overburden, which is soil and rock of normal radium-226 concentrations, and then a layer known as the leach layer, which has a high radium-226 concentration, before the ore is exposed. After ore removal the overburden and leach layers are replaced but in a haphazard fashion resulting in areas where the leach layer is at the surface on top of the original overburden. Buildings constructed on this reclaimed land often had elevated radon-222 concentrations in indoor air and remedial action was introduced in Florida along similar lines to that taken in Colorado<sup>(24,25)</sup>. The experience with uranium mill tailings and the reclaimed phosphate lands high-lighted the degree of influence that the radium-226 concentration in the soil beneath the building can have on the radon-222 concentration in air within the building.

Data has also been published on the radium-226 and thorium-232 concentration in common building materials in the USSR. This illustrates the variability of concentration in similar finished articles manufactured in different locations<sup>(26)</sup>. A selection of these results is given in Table 3 for illustration.

Table 3  
Specific activity of radium-226 and thorium-232 in materials  
from the USSR

Material	Locality	Number of specimens	Specific activity (Bq kg <sup>-1</sup> )	
			Thorium-232	Radium-226
Red brick	Leningrad	14	41	60
	Kazakhstan	23	27	54
	Nalchik	8	40	54
	Novgorod	8	54	59
	Cheliabirsk	2	24	40
Lime brick	Kazakhstan	6	12	19
	Kirghijia	1	38	52
	Kalinin	1	13	14
	Leningrad	2	10	27
	Zaporoshic	3	22	33

### 3.2 Thorium-232 and radium-226 specific activities in a selection of building materials used for radon-222 emanation studies carried out for this report

In the studies reported here the major emphasis has been on determining the emanation rate of radon-222 from building materials commonly used in the UK. However, it is of interest to determine that fraction of the radon-222 that is formed within the material that escapes from the surface to the surrounding air. This necessitates determining the radium-226 specific activity of a selection of materials. The method of determination of the radium-226 specific activity and that of thorium-232 is given in Appendix B. Table 4 presents the specific activity results obtained for single specimens of each type shown. Appendix C gives detailed data, where available, on the constituents of the building materials examined.

Table 4  
Specific activities of a selection of building materials  
used for radon-222 emanation studies

Material		Specific activities (Bq kg <sup>-1</sup> )	
Type	Main constituent	Thorium-232	Radium-226
SL20 brick	Silica	14.8 ± 0.7	32.9 ± 0.9
F54 brick	Flint	51.6 ± 1.6	69.6 ± 1.6
Kirton brown brick	Clay	51.2 ± 1.8	84.2 ± 2.7
Kirton brown brick	Clay	39.5 ± 1.3	63.7 ± 1.9
Dapple light brick	Clay	59.0 ± 1.9	77.4 ± 2.9
Otterham second hardstock brick	Clay	52.1 ± 1.3	31.1 ± 0.6
Severn valley orange multi-brick	Clay	36.0 ± 0.9	56.9 ± 1.9
Ibstock red rustic multi-brick	Clay	51.0 ± 1.6	76.5 ± 2.5
Ludlow crushed granite brick	Granite in matrix	6.2 ± 0.3	22.0 ± 0.5
Fyfe-stone block	Granite	54.7 ± 2.3	49.0 ± 2.7
Fyfe-stone block	Granite	54.7 ± 2.4	50.0 ± 2.4
Insulating block	PFA	170.0 ± 3.0	77.0 ± 2.0
Block	LECA	55.0 ± 1.2	92.4 ± 2.3
Block	LECA	58.5 ± 2.3	96.4 ± 2.3
Brick	Oil shale	42.9 ± 1.9	76.3 ± 1.9
Block	Concrete	42.1 ± 0.7	17.7 ± 0.7

PFA = pulverised fuel ash; LECA = light expanded clay aggregate.

For the six clay bricks measured (see Table 4) the mean radium-226 specific activity is  $65.0 \pm 19.3$  (unweighted)  $\text{Bq kg}^{-1}$  which is close to that derived for clay bricks from all sources in Tables 1 and 2.

Although the number of specimens shown in Table 4 is limited, the clay brick results illustrate that the variability in specific activity between specimens of the same type (the two Kirton brown bricks) is of the same order as that between disparate specimens. The results obtained for the two Fyfe-stone blocks and the two LECA blocks are remarkably consistent. This presumably indicates a more homogeneous composition of the constituents of these blocks than is the case for the clay bricks (at least for the Kirton brown). The Kirton brown brick has as its main constituent Keuper Marl, presumably extracted from an area close to the brick works, and the approximately 25% difference in radium-226 specific activity of the two specimens is not readily explained. However, the radon-222 emanation studies on these bricks supported the difference in specific activities, as will be shown in Section 4.2.

#### 4. EMANATION OF RADON-222 FROM BUILDING MATERIALS AND SOILS

The limited data on the emanation of radon-222 from building materials that have been reported elsewhere are given in Section 4.1. It is now widely believed that the soil beneath the building is, in most cases, the major source of radon-222 within the building. For this reason some data is also presented in this section on values of the radon flux from the ground that have been measured world-wide. The results of radon emanation studies carried out at NRPB are summarised in Section 4.2; a complete presentation of the data together with details of the composition of the materials, where available, is given in Appendix C.

##### 4.1 Reported values from the literature of the emanation of radon-222 from building materials and soils

There have been four reports world-wide on the measured values of radon emanation rates from building materials. The most extensive to date is the data obtained in Denmark and presented in Table 5<sup>(27)</sup>. Table 5 gives emanation rates for each material in terms of per unit area, per unit volume and per unit mass. These parameters will also be used to present the results of the studies carried out at NRPB. It must be stressed, however, that the surface area of some building materials is almost impossible to define. This is true of a number of hand-made bricks with cracked or fissured surfaces, which also affects the assessment of volume but to a lesser extent.

Each measurement reported in Table 5 represents a measurement on a single specimen. However, an attempt was made to examine the effects of geometry on the radon emanation rate for a given material. The two materials tested in this way were the Danish light-weight concrete and the clay bricks. The two light-weight concrete specimens were sliced from a large concrete block, such that the two specimens measured had the same composition but different surface area to volume

Table 5  
Emanation rates of radon-222 from building materials  
from Denmark and Sweden

Material	Emanation rates		
	Bq m <sup>-2</sup> s <sup>-1</sup> x 10 <sup>-6</sup>	Bq m <sup>-3</sup> s <sup>-1</sup> x 10 <sup>-3</sup>	Bq kg <sup>-1</sup> s <sup>-1</sup> x 10 <sup>-6</sup>
Ordinary concrete, gravel and sand from the sea, Danish deposits	350	12	5.0
Ordinary concrete, gravel and sand from pits, Danish deposits	280	9.3	4.2
Ordinary concrete, composition unknown	330	11	4.7
Ordinary concrete, composition unknown	390	12	4.8
Light-weight concrete, Swedish origin, alum-shale, old type	8600	240	440
Light-weight concrete, Swedish origin, alum-shale, new type	3000	96	150
Light-weight concrete, Danish origin, clay based	67	2.2	3.0
Light-weight concrete, Danish origin, clay based.	44	2.3	3.0
Expanded clay concrete, LECA	46	1.6	2.5
Solid clay brick	4.8	0.33	0.17
Clay bricks with cavity	2.0	0.33	0.17
Chipboard	0.71	0.13	0.19
Fibreboard	1.1	0.19	0.23
Gypsum board	1.3	0.23	0.23

ratios. The clay bricks were of the same material, with one having cavities and the other being solid. The results for these specimens are repeated for convenience in Table 6 together with their respective surface area to volume ratios.

It appears from this limited data that the emanation rates per unit mass or volume are less influenced by the geometry than is the emanation per unit surface area. It is interesting to note that the most consistent results are from different bricks of ostensibly the same composition, rather than the two concrete specimens hewn from the same block. As is shown in Section 4.2 and Appendix C, different specimens of the same building material can have emanation rates that differ by up to a factor of two, and perhaps the agreement in Table 6 is fortuitous, with the concrete results being more representative.

Table 6

The effect of geometry on the radon-222 emanation rate

Material	Surface/ volume ratio (m <sup>-1</sup> )	Radon-222 emanation rates		
		Bq m <sup>-2</sup> s <sup>-1</sup> x 10 <sup>-6</sup>	Bq m <sup>-3</sup> s <sup>-1</sup> x 10 <sup>-3</sup>	Bq kg <sup>-1</sup> s <sup>-1</sup> x 10 <sup>-6</sup>
Light-weight concrete	33	67	2.2	3.0
Light-weight concrete	52	44	1.6	2.5
Solid clay brick	67	4.8	0.33	0.17
Cavity clay brick	161	2.0	0.33	0.17

Stranden and Berteig have measured the radium-226 concentration in Norwegian building materials and investigated the radon-222 emanation rates for different thicknesses of material formed into wall sections<sup>(28)</sup>. Their results are presented in Table 7.

Table 7

Radium-226 concentration and radon-222 emanation rates for  
some Norwegian materials

Material	Radium-226 (Bq kg <sup>-1</sup> )	Thickness (cm)	Radon-222 emanation rate (Bq m <sup>-2</sup> s <sup>-1</sup> 10 <sup>-6</sup> )
Concrete	33	20	4600
Concrete	33	150	8600
Brick	56	20	3100
Brick	56	70	5600
LECA	81	10	3200
LECA	81	20	5900

It should be noted that the data in Table 7 for wall sections indicate emanation rates per unit area two orders of magnitude greater than those for isolated specimens of building materials (eg, brick) given in Table 5. Although the emanation rate per unit area of a wall section is expected to be higher than that for an isolated element of the building material, an increase of this magnitude is unlikely.

Measurements of the radium-226 concentration in building materials have been carried out in Poland and the emanation power of the materials calculated<sup>(29)</sup>. The emanation power is the ratio of the number of atoms of radon-222 that escape from a material into the surrounding air per unit time to the total number of radon-222 atoms formed within the material per unit time. The emanating power is often expressed as a percentage and is presented thus for the Polish results in Table 8.

Table 8 reveals that the radium-226 specific activity of a material is a poor indicator of its potential as a source of radon-222 in room air. For the



Table 8  
Emanation powers and radium-226 specific activity  
of some building materials

Material	Number of specimens	Radium-226 specific activity (Bq kg <sup>-1</sup> )		Emanating power (%)	
		Range	Mean	Range	Mean
Fly ash	33	63-140	96	0.2-1.16	0.54
Slag	11	37-130	67	0.24-1.53	0.70
By-product gypsum	4	26-736	26	3.50-21.3	3.5 <sup>1</sup>
			700		12.9 <sup>2</sup>
			736		21.3 <sup>3</sup>
			580		19.7 <sup>4</sup>
Red brick	3	19	19	2.20-5.1	3.2
Silica brick	3	7.4-15	10	0.80-16.3	6.8
Cement	4	< 9-26	18	0.80-8.5	4.6
Soil	5	3.7-19	11	12.10-50.1	28.9

<sup>1</sup>Kola (apatite).

<sup>2</sup>Florida (phosphorite).

<sup>3</sup>Morocco (phosphorite).

<sup>4</sup>Togo (phosphorite).

three specimens of red brick tested there is more than a factor of two between the lowest and highest emanating power, yet all had the same specific activity. In the case of the silica bricks, the specimen having an emanating power of 16.3% had a specific activity of 7.4 Bq kg<sup>-1</sup> whereas the red brick of emanating power 5.1% had a specific activity of 19 Bq kg<sup>-1</sup>. Thus, for equal weight bricks, the radon emanation from the silica brick would be 24% higher than that of the red brick, whereas the specific activity of the silica brick is only 39% that of the red brick. Although the higher the emanating power the stronger the material acts as a source of radon-222, it should be remembered that a low emanating power means that more of the radon-222 is retained within the material and this will increase the gamma-dose rate from the material.

It has been stated that in many (if not most) cases the predominant source of radon-222 within buildings is the soil and rock beneath the structure. This has proved a particular problem in the case of buildings on the reclaimed phosphate lands of Florida and in the uranium mining communities of Canada. In the latter case, dwellings (invariably with basement) have been built over rock containing uranium ore bodies. If building is contemplated on land that might contain elevated uranium concentrations, certain forms of construction can minimise the influence of the soil on indoor radon-222 concentrations. Before a decision on the form of construction is taken measurements of the flux of radon-

222 from the soil would be advisable. Extensive measurements of the radon-222 flux from soils world-wide have been carried out and the results collated by Wilkening et al<sup>(30)</sup>. These values are given in Table 9.

Table 9  
Radon-222 flux from soils world-wide

Location	Soil group	Radon-222 flux (mBq m <sup>-2</sup> s <sup>-1</sup> )
<u>Austria</u>		
Graz	Mountain	20, 9.2
Innsbruck	Mountain	8.6, 18
<u>France</u>		
Saclay	Podsol	15, 14
<u>Germany</u>		
Aacken	Podsol	17
<u>Ireland</u>		
Dublin	Podsol	27
<u>Japan</u>		
Osaka	Latosol	3.4, 8.8
<u>Phillipines</u>		
Manilla	Latosol	11
<u>USA</u>		
Socorro, NM	Desert	36
Yucca Flat, NV	Desert	18
Lincoln, MA	Podsol	50
Champaign County, IL	Chernozem	52
Argonne, IL	Chernozem	21
<u>USSR</u>		
Kirov	Podsol	15
Moscow	Podsol	3.8
Central European	Podsol/	6.9
Territory	chernozem	
Southwest Kazakhstan	Desert	5.0
The North, Murmansk	Podsol	3.8
The Caucasus	Mountain/	11
	chernozem	
Middle Asia	Mountain/	18
	desert	
South Urals	Mountain/	11
	podsol	

The authors are not aware of any extensive measurements of the radon-222 flux from soil in Great Britain, but a few measurements at Chilton, where the soil is of the rendzena type, indicate a value for this area of 27 mBq m<sup>-2</sup> s<sup>-1</sup>.

The total number of measurements considered by Wilkening et al was 994. From these they conclude that the average radon-222 flux is 16 mBq m<sup>-2</sup> s<sup>-1</sup> with a range of 0.21-52 mBq m<sup>-2</sup> s<sup>-1</sup>. This indicates that the total radon-222 flux from the world's land areas is 2 10<sup>12</sup> Bq s<sup>-1</sup>.

#### 4.2 Radon-222 emanation rates and emanating powers measured at NRPB

Some 165 specimens of building materials have been evaluated for their emanation of radon-222. In the case of bricks of different manufacture, five specimens of each type were usually measured but occasionally only four. Other materials, including bricks of unusual composition, were available as single specimens only. A full list of all emanation studies is presented in Appendix C, together with details of the constitution of those materials where the manufacturers have been kind enough to supply the information.

In this section a broad picture of the emanation rates from bricks and other materials is presented, and details are given of the radon emanation rates of those materials whose radium-226 concentration has been determined. The methods of determining the radium-226 concentration and the radon-222 emanation rates of a material are given in Appendix B.

The results of the emanation rate studies demonstrate quite large intra-type variation, as well as the expected inter-type variation. Thus, for a sample of five specimens of the same brick, it is not unusual to find a difference of a factor of two between the specimen with the lowest emanation rate and that with the highest.

Table 10 gives an overall picture of the radon-222 emanation rates for broad classes of materials, but a number of results are given of single specimens of unusual manufacture to indicate the range of values that might be encountered. In this report, sample is used to indicate a type of product, such as Kirton brown or SL20 bricks, and specimen refers to an individual article, such as a single brick.

Table 10 indicates that silica bricks are considerably stronger sources of radon-222 than are clay bricks, but in both cases a wide range of values is found. The Fyfe-stone block composed of granite chippings has the highest radon emanation of any material tested during this programme, but this material is not commonly used within buildings. The use of less conventional constituents in brick making can have quite different effects on the radon emanation properties of the final product. The Ludlow crushed granite brick, as its name implies, has crushed granite as a major ingredient and must have an open matrix, as will be shown later. This brick has a high emanation rate but it is less than half the value of that of the brick containing oil shale. Conversely, the use of colliery shale has produced a brick with a low emanation rate, towards the bottom of the range for clay bricks of more usual constitution.

The results from this programme of work are in reasonable agreement with the results from Denmark given in Table 5.

From the emanation rates and a knowledge of the radium-226 concentration in the specimen the emanating power can be determined. Table 11 gives the emanation rates and radium-226 concentrations, and hence the emanating power for those materials listed in Table 4.

Table 10

## Radon-222 emanation rates of some building materials

Material type	No. of samples	No. of specimens	Radon-222 emanation rates of some building materials					
			Bq m <sup>-2</sup> s <sup>-1</sup> x 10 <sup>-6</sup>		Bq m <sup>-3</sup> s <sup>-1</sup> x 10 <sup>-3</sup>		Bq kg <sup>-1</sup> s <sup>-1</sup> x 10 <sup>-6</sup>	
			Mean	Range	Mean	Range	Mean	Range
Clay brick	18	86	13	3.6 - 30	1.0	0.32 - 1.8	0.56	0.15 - 1.0
Silica brick	4	27	80	33 - 130	5.6	2.2 - 9.2	3.2	1.1 - 5.5
Fyfe-stone block <sup>1</sup>	2	6	590	510 - 790	22	19.0 - 28	9.3	8.8 - 9.5
Concrete block <sup>1</sup>	3 <sup>2</sup>	9	120	50 - 230	4.3	1.9 - 7.6	2.2	1.2 - 3.8
Light-weight concrete block	1	3	12	6.8 - 15	4.1	2.3 - 5.1	4.7	2.6 - 5.7
LECA block	1	2	30	23 - 36	1.0	0.77 - 1.2	1.2	0.90 - 1.4
Ludlow crushed granite brick	1	3	130	88 - 180	7.8	5.2 - 10	3.6	2.4 - 5.2
Standard brick containing colliery shale	1	1	5.6	-	0.41	-	0.24	-
Standard brick containing oil shale	1	1	290	-	17	-	11.1	-

<sup>1</sup>Where results are given for a small number of samples the range given is that for all specimens.<sup>2</sup>Each sample was of a different thickness.

Table 11  
Emanating Power of a selection of building materials

Material	Radium-226 concentration (Bq kg <sup>-1</sup> )	Radon-222 emanation rate (Bq kg <sup>-1</sup> s <sup>-1</sup> 10 <sup>-6</sup> )	Emanating power (%)
SL20 silica brick	32.9	1.6	2.3
Kirton brown clay brick	84.2	0.69	0.39
Kirton brown clay brick	63.7	0.39	0.29
LECA block	92.4	0.90	0.46
LECA block	96.4	1.4	0.69
LBC dapple light clay brick	77.4	0.69	0.42
F54 buff flint and lime brick	69.6	1.2	0.82
Ludlow crushed granite brick	22.0	5.2	11
Grey Fyfe-stone block	49.0	12	12
Pink Fyfe-stone block	50.0	8.8	8.4
Otterham second, hard stock clay brick	31.1	0.96	1.5
Insulating block containing PFA	77.0	3.6	2.2
Severn Valley orange multi- clay brick	56.9	0.43	0.36
Ibstock red rustic clay brick	76.5	0.56	0.35
Brick containing oil shale	76.3	11	6.9
Concrete block	17.7	1.4	3.8

It is apparent that the radium-226 concentration alone is not a reliable indicator of the radon-222 emanation rate. The Ludlow crushed granite brick has a relatively low radium concentration but, being relatively porous, a large fraction of the radon formed within the brick can escape to the surface. The Kirton brown clay brick has a radium concentration that is quite high, but is a compact product from which little radon escapes. All silica bricks measured during this programme yielded higher radon emanation rates than clay bricks of traditional manufacture. From the limited data presented here, this is apparently due to their higher emanating power rather than an elevated radium concentration. It is of interest to note that where bricks are to be used to build an enclosure within which the gamma-ray dose rate is to be minimised, the bricks used should prefer-

ably have both a low radium concentration and a high emanating power. This follows from the fact that the gamma-rays associated with radium-226 are actually those from the decay products of radon-222, ie, lead-214 and bismuth-214. Only if the radon, and hence its decay products, are retained within the material will the material produce a gamma-dose rate due to these nuclides.

Emanation rates have been given correct to two significant figures only. This reflects the repeatability of a measurement which is seldom better than 10% as, the emanation rate from a given specimen is not time invariant (see Appendix B, section 2).

#### 4.3 Radon-222 emanation from wall sections

The emanation of radon-222 from a building material, such as an isolated brick, will not have the same value when that material is incorporated in a segment of a building such as a wall. For a brick in the centre of a wall there will be little diffusion of radon in the plane of the wall, as here the concentration gradients will be small. The major emanation rate will be in the direction perpendicular to the wall, and hence the emanation rate per unit area for the face of the brick exposed in the wall will be higher than the value measured for all faces of the brick when the brick was isolated. The actual emanation rate increase will depend upon the position of the brick within the wall and will range from a value close to that obtained when the brick was isolated to more than three times that value. This would be true of a wall comprising a single stack of bricks with opposing faces exposed to the air. In practice, walls of brick or other material internal to a building are often rendered with cement and plastered. This will attenuate the radon flux from the wall material while adding a component of its own to the final emergent flux.

Attempts have been made to measure the radon flux from wall sections by sealing a can to the wall in which radon escaping from the wall is trapped. A method of achieving an air-tight seal to the wall without marking the wall has not been found and only limited data are available. These were obtained from office walls at NRPB and in one dwelling. For a partition wall of breeze-block construction, rendered and plastered, two measurements gave values of  $6.6 \times 10^{-6}$  and  $6.3 \times 10^{-6} \text{ Bq m}^{-2} \text{ s}^{-1}$ , respectively. Measurements on the wall of the dwelling, where the can was sealed to a rendered, plastered and papered brick wall, produced emanation determinations of  $1.8 \times 10^{-5}$ ,  $2.4 \times 10^{-5}$ ,  $2.0 \times 10^{-5}$  and  $1.7 \times 10^{-5} \text{ Bq m}^{-2} \text{ s}^{-1}$  over four successive measurement periods. Whereas in the office measurement it was possible to flush the collecting can with radon-free air, this was not possible in the dwelling, and thus the initial air in the collecting can contained radon at a concentration typical of that of the room (less than  $10 \text{ Bq m}^{-3}$ ).

Neither of the walls tested appear to have a high emanation rate for radon and more information needs to be gathered. McLaughlin and Jonassen measured the emanation rate from the concrete walls of a basement and found the rate to be

$8.0 \pm 2.2 \cdot 10^{-4} \text{ Bq m}^{-2} \text{ s}^{-1}$  which they considered to be high<sup>(31)</sup>. The most likely explanation for the higher emanation rates reported by McLaughlin and Jonassen is that the walls, being part of a basement, have soil against the outside and some of this radon flux is due to radon diffusing in from the soil. The Norwegian data given in Table 7 does not appear to be representative of conditions for wall sections in constructed buildings. Accepting that an emanation rate of  $8 \cdot 10^{-4} \text{ Bq m}^{-2} \text{ s}^{-1}$  is higher than average, but using this figure to calculate the radon concentration in a room where every surface emanates at this rate, a figure towards the upper limit of that which can be attributed to the building materials will be obtained. For a room of surface to volume ratio  $1.8 \text{ m}^{-1}$  this yields a radon-222 concentration, at a ventilation rate of one air change per hour, of  $5.2 \text{ Bq m}^{-3}$  ( $0.14 \text{ pCi l}^{-1}$ ). This is a lower concentration than is experienced in most dwellings under these conditions and demonstrates that in most situations, while the building materials cannot be neglected as a source of radon-222 in room air, other sources dominate in many circumstances. This point will be emphasised in Section 5.

#### 4.4 The effect of coating building materials as a method of reducing radon emanation

When a radon atom is formed by the decay of its radium-226 parent which is embedded within a building material, the radon atom will be free to migrate to the surface only if it is formed close to a pore space. The diffusion coefficient of radon in the crystalline material is some 20 orders of magnitude smaller than that in air. Thus radon formed within the crystalline material will decay before it reaches the pore spaces. Radon formed close to, or on, the walls of a pore space will be free to diffuse to the surface. Generally, the higher the porosity of the material the greater the number of radon atoms likely to be formed near a pore space, and hence the higher the radon emanation and emanating power. It must be noted that porosity alone is not a reliable indicator of emanating power as porous materials can be loosely classed as being of two types. Porosity is simply defined as the ratio of the pore volume to the total volume of the material. If a material has pores consisting of isolated cells, radon formed within these pores has to cross solid material to reach the surface and the emanation will be low. A second material, having the same porosity but in which the pore spaces are linked by air filled channels, will have a much higher emanation rate if the amount of radon reaching the pore spaces is precisely the same as in the former case. One approach to reducing the emanation of radon from a material is to coat the material with a substance that will clog the pore spaces near the surface and form a layer at the surface in which radon has a low diffusion coefficient.

A problem with many coating materials is that of durability. Any measures taken to reduce the emanation must be effective for many years to warrant the attempt. Muburak<sup>(20)</sup> applied gloss paint to granite and achieved an immediate

reduction in emanation rate of 57%. However, no follow up measurements are reported. The authors have applied a variety of paints to bricks with mixed results. These results are summarised in Table 12. In each case, two coats of the relevant paint were applied to the brick.

Table 12  
Effects of common paints applied to silica bricks

Paint	Emanation rate (unpainted = 100%)	
	At 3 days	At 8 weeks
Oil based primer <sup>1</sup>	-	97
Oil based undercoat	90	90
Oil based gloss	52	102
Emulsion	93	not tested

<sup>1</sup>At 3 days the radon emanation from this sample was apparently nearly three times that of the unpainted specimen. It is thought that this spurious result was due to paint solvent entering the measuring system.

The early promise shown by the oil based gloss paint did not persist due to cracking of the coating. The undercoat is promising having endured for eight weeks, although the reduction is marginal. Much longer testing periods are needed and are planned.

A series of paint applications were given to one silica brick with emanation rate determinations taking place between coats of paint. The results for this specimen are given in Table 13.

Table 13  
Variation in radon-222 emanation from one silica brick following sequential paint applications

Treatment	Emanation
Unpainted	100
+ 1 coat oil based primer	76
+ 1 coat oil based primer	94
+ 1 coat oil based under-coat	105
+ 1 coat oil based gloss	54

The results following the second and third coats of paint are not significantly different from the unpainted value, whereas that after the first coat is significant, but apparently not durable. The reduction in emanation rate following the application of gloss paint is real, but will have to be assessed periodically to determine how durable this effect is.



For two other silica bricks, three coats of paint were given to each brick; in one case undercoat and in the other gloss paint. The emanation rates were measured after each application of paint and the results were as shown in Table 14.

Table 14  
Effect of progressive coats of paint on the emanation rate  
of silica bricks

Paint	Emanation rate (unpainted = 100%) after applying coat		
	1	2	3
Oil based undercoat	93	95	97
Oil based gloss	88	88	47

In this case the emanation from the brick painted with undercoat is not significantly different from that for the unpainted brick. Again the gloss paint shows promise, but its durability cannot be determined adequately without periodic measurements of the emanation rate over several years. The brick painted with gloss paint, reported in Table 14, was remeasured 6 weeks after the above measurement and the emanation rate was 51% of the unpainted value. This is not statistically significantly different from the 47% first reported.

The most extensive studies of paint materials as radon barriers have taken place in the USA. In areas where uranium mill tailings have been used as back-fill material around buildings and as an aggregate for concrete, attempts have been made to limit the ingress of radon into the structure by applying coating materials to internal walls and floors. As mentioned earlier, it is expected that the gamma-dose rate will increase from materials from which the escape of radon is reduced or prevented. In most cases it is unlikely that the gamma exposure would increase by more than 10-20%<sup>(32,33)</sup>. The most successful sealants are based on epoxy resins and are the type of material used to produce hard wearing waterproof wall and floor surfaces in buildings, such as airport buildings. A perfect seal against radon emanation was achieved by applying four coatings of epoxy resins with various additives and fillers. The coats were applied at daily intervals and the final thickness of the coat was 4.1 mm<sup>(34)</sup>. This treatment would seem more suitable for floors and is relatively expensive. Although this provides a hard and durable finish it is not known whether micro-cracks appear in the resin due to small movements of the underlying concrete. Other workers using epoxy paint have reported reductions in emanation to 51-41% of the untreated value, depending upon whether or not a filler is used, and a reduction to 68% with latex paint<sup>(35)</sup>.

The first concern about the health hazards arising from the inhalation of the short-lived decay products of radon-222 occurred in the uranium mining industry. Within the mines the standard method for reducing the radon concentration has been to increase the ventilation by pumping fresh air from above ground through the mine and venting the radon laden air to the outside. This can be very effective, but in deep mines the expense is high and in some areas of the mine, such as blind galleries, it is not easy to ensure air movement. In such areas an alternative method of reducing the radon concentration is desirable and one such method is to coat the rock face with a material that will reduce the radon emanation. The United States Department of the Interior, Bureau of Mines have carried out extensive studies of polymeric materials for sealing radon into walls<sup>(36)</sup>. The materials used had to provide a cure under the adverse conditions in a mine of high temperature and very high humidity. The materials found to be successful as radon barriers were a two-component water-based epoxy paint which reduced the emanation of radon to 20% of the untreated value; a furan paint that initially reduced the emanation to 2% and at nine months after application the emanation was still only 5% of the untreated value; a one-component unsaturated polyester paint which again reduced the emanation to 5% after 9 months; and other paint materials which were not commercially available also reduced the emanation to between 20 and 10%. The cost of application of these materials is probably below £5 m<sup>-2</sup>. In those cases where high radon concentrations within buildings results from the inclusion of material of relatively high uranium concentrations in the aggregate of the construction concrete, the coatings offer a remedial strategy which is reasonably low in cost and causes the least disturbance to the normal use of the building.

#### 4.5 The use of membrane materials to reduce the ingress of radon

The radon-222 concentration in the air within buildings can very rarely be supported by the radium-226 content of the building materials surrounding the room. The only other source of sufficient magnitude is the soil and rock beneath the structure and this has proved, in most cases, to be the dominant source. This point is illustrated by a reported measurement series in Chicago, USA. If the building materials are the major source of radon-222 within dwellings then wood-frame dwellings would be expected to support radon concentrations considerably lower than those in masonry dwellings, as wood has a low radium-226 concentration. Measurements in houses in Chicago generally gave radon-222 concentrations in the range 5-160 Bq m<sup>-3</sup>. However, in one wood-frame house the radon-222 concentration in the bedroom was 960 Bq m<sup>-3</sup>. The source of this radon was traced to the unpaved crawl space (solum) under the kitchen floor where the emanation from the soil was found to be 0.27 Bq m<sup>-2</sup> s<sup>-1</sup><sup>(37)</sup>. This figure is more than two orders of magnitude higher than the strongest emanating material listed in Table 10 (the Fyfe-stone block having a mean emanation rate 5.9 10<sup>-4</sup> Bq m<sup>-2</sup> s<sup>-1</sup>) and is an order of magnitude higher than the value for typical soils.

This is an extreme example of the effect of radon ingress from the ground, but less dramatic, yet significant, results have been documented world-wide. As a rather rough guide, it has been suggested that for ground-floor rooms or single family dwellings, 50% of the radon within the building originates from the ground beneath the dwelling, 25% from the structural elements and 25% from the outside air<sup>(21)</sup>. The precise mix will vary quite widely depending upon the local geology and the building materials employed.

In dwellings with a suspended floor and well-ventilated solum the effect of radon from the ground is greatly reduced, except under very still air conditions. In fact, when building on land that is known to be contaminated with radium this is the preferred method of construction<sup>(38)</sup>. A more common form of construction for dwellings built recently in the UK is that of a concrete slab or raft (referred to in the American literature as slab on grade). Radon, being a noble gas, reacts chemically with materials only under extreme conditions and as concrete is porous radon will diffuse through it more readily than more reactive gases. The effective diffusion coefficient for radon through intact concrete has been found to be  $1.69 \cdot 10^{-5} \text{ cm}^2 \text{ s}^{-1}$  for concrete of porosity 0.05, and  $3.08 \cdot 10^{-5} \text{ cm}^2 \text{ s}^{-1}$  for concrete of porosity 0.25<sup>(39)</sup>. The relaxation length of a material is defined as that thickness of material, through which a radioactive gas is diffusing, that reduces the flux to a value of  $1/e$  of the initial value. The reduction in flux results from the finite time taken to diffuse this distance, during which some of the gas decays. The relaxation lengths for radon in concrete for the two values of diffusion coefficient given above are 12.7 and 7.6 cm, respectively. A typical value for the relaxation length of radon-222 in concrete may be taken as 10 cm. This might indicate that a 10 cm slab of concrete (containing no radium-226) laid on soil would reduce the radon-222 flux from the soil to 37% of the value from the bare soil. However, the simple application of the relaxation length concept only applies within a homogeneous medium. In the case considered here there is a discontinuity in the medium at the soil-concrete interface. When the concrete slab is laid on the soil the boundary conditions at the soil surface change due to the different radon-222 diffusion coefficients in concrete compared to that in air. A 10 cm slab of concrete (crack free and devoid of radium-226) placed on soil will result in a radon-222 flux through the top surface of the slab that is of the order of 1% of the value of the flux from the uncovered soil (see Appendix B, Section 6). Taking the mean value for the radon-222 flux from soil, given in Section 4.1, of  $1.6 \cdot 10^{-2} \text{ Bq m}^{-2} \text{ s}^{-1}$ , the flux emerging from the concrete slab would be about  $1.6 \cdot 10^{-4} \text{ Bq m}^{-2} \text{ s}^{-1}$  if the concrete contained no radium-226, and this figure is an order of magnitude larger than the emanation rate of clay bricks determined in the work reported here (see Table 10). In practice the slab will contain some radium-226 and this will produce an additional component to the emergent radon-222 flux. Thus, even with the large reduction in radon-222 flux effected by an uncracked

concrete slab, the subjacent soil and concrete slab form a significant source of radon-222 in room air above the slab.

In practice, concrete slabs do not remain intact and cracking will occur, although many will be so small that they are difficult to detect. As soon as a crack appears, however small, the crack (being air filled) provides an easy route for radon-222 from the soil to the room above the slab. A model to determine the effects of cracks in concrete has been developed<sup>(40)</sup>. In the worst case considered, a 0.5 cm crack every 0.5 m of slab, the total flux through the concrete slab is 28% of the value from an area of soil equal to that of the slab. At the other extreme, a 1 cm crack every 2 m of slab, the radon flux through the crack is calculated to be about 10% of the total flux from the same area of soil as the total slab. In the model the concrete is regarded as impermeable, as the diffusion coefficient of radon in concrete is four orders of magnitude smaller than that in air. In practice, some radon will diffuse through the concrete as well, as outlined above.

When construction of the concrete slab is undertaken it is usual to lay a 500 or 1000 gauge polythene membrane in the slab to retard moisture. Radon will diffuse through most plastics, and during this programme of work the diffusion coefficient of radon in polythene (using 1000 and 500 gauge material) and in polyvinylchloride has been measured. For polythene the diffusion coefficient was  $3.3 \times 10^{-7} \text{ cm}^2 \text{ s}^{-1}$  and for PVC  $1.1 \times 10^{-7} \text{ cm}^2 \text{ s}^{-1}$ .

An intact membrane of 1000 gauge polythene would, if sandwiched in a 10 cm concrete slab, reduce the radon flux from the top of the slab to 84% of the value without the membrane, rather than to only 93.8% as predicted by the relaxation length concept (see Appendix B, Section 5). Other materials with lower diffusion coefficients could be effectively used to reduce the radon flux. Polyamide is reported to have a diffusion coefficient for radon-222 of  $< 10^{-11} \text{ cm}^2 \text{ s}^{-1}$ <sup>(41)</sup>. A 1000 gauge membrane of this material would reduce the radon flux to less than 0.001% of the value without the membrane.

## 5. AIRBORNE CONCENTRATIONS OF RADON-222, RADON-220 AND THEIR DECAY PRODUCTS

### 5.1 Reported measurements outside the UK

Radon had been known to exist in ambient air for several decades before any serious studies of the concentration of radon-222 in indoor air were undertaken. The seminal study in this field is that of Hultqvist in Sweden in 1956<sup>(42)</sup>. He noted that the concentration of radon-222 in indoor air was often an order of magnitude higher than that in the open air. He also noted that the concentration of radon-222 in air within buildings is subject to rapid variation with excursions larger than those noticed in outside air, and he proposed a method of determining the rate of production of radon-222 within room air, which is discussed in Section 5.2. Radon-222 formed in soil diffuses to the surface, and in the open is rapidly diluted by the vast volume of the atmosphere. Radon-222 diffusing into room air is not so readily diluted; the extent of dilution

being critically dependent upon the ventilation rate. In a hermetically sealed room the concentration of radon-222 would increase and approach, asymptotically, a value at which the rate of production of radon-222 within the room was equal to the rate of its radioactive decay. In a room with a ventilation rate in excess of 15 air changes per hour the concentration will be little different from that in outside air (providing the room does not contain an unusually strong source of radon-222). For intermediate ventilation rates the radon-222 concentration will lie between these limits and will be inversely proportional to the ventilation rate for rates exceeding 0.1 air changes per hour.

Reports of measurements of the radon-222 concentration and the concentrations of radon-222 decay products in room air in many countries have appeared in the literature over the past quarter of a century. Unfortunately, there is no uniform method of presenting the data and this makes comparisons difficult in some cases. The difficulties are compounded by the vast range in study size, from three rooms<sup>(43)</sup> to one of 9999 dwellings<sup>(44)</sup>. Table 15 lists some of the reported measurements of the concentration of radon-222 and its decay products, both inside buildings and in the open air. In a few cases some data on the concentrations of radon-220 found are also given.

For all reported measurement programmes covering more than 20 buildings or rooms the distribution of measured radon-222 or radon-220 decay products concentrations is positively skewed, with the mean considerably greater than the median value. In most studies the mean radon-222 concentration is in the range 30–100 Bq m<sup>-3</sup>. The results given in Table 15 for Norway are of interest in that the radon-222 concentration (for similar conditions of ventilation and underlying rock) would be expected to be greatest in concrete structures and least in those of wood. The anomalous situation whereby the wood construction dwellings apparently have a higher mean radon-222 concentration than those of brick construction may be due to the relatively small and unequal numbers in each group.

Although the number of measurements reported from Schenectady, USA are very small the large difference in radon-222 concentration between basements and first floor rooms is substantiated by many other measurements. The radon-222 concentration in basements let into soil overlying uranium ore bodies have been found to be much higher than the concentration in rooms at higher elevation. This problem was most pronounced in dwellings in the mining communities of Canada and led to a programme of remedial action to reduce the radon-222 concentrations in these buildings. Once again the uranium concentration in the subsoil was the dominant source of radon-222 in the basement air. Here, five room surfaces are surrounded by soil from which radon can diffuse through the floor and walls into basement air. Basements are not a common feature of UK housing, but the few measurements that have been made confirm that a higher radon-222 concentration is invariably found in the basement of a building than elsewhere in that building.

Table 15  
Selected measurements of the concentrations of radon-222, radon-220 and their decay  
products in air within buildings and in the open air

Country	Details	Reference
Austria (No. of rooms not stated)	Mean radon-222 concentration in living rooms in the range 28 - 115 Bq m <sup>-3</sup> Maximum radon-222 concentration 276 Bq m <sup>-3</sup> Mean radon-220 concentration in room air in the range 4 - 41 Bq m <sup>-3</sup>	45
	Median radon-222 concentration indoors with geometric standard deviation (GSD) 15 Bq m <sup>-3</sup> 2.6 Median radon-222 concentration outdoors 8.1 Bq m <sup>-3</sup> with GSD 2.7 Minimum concentration of radon-222 in both situations 1.9 Bq m <sup>-3</sup> Maximum indoor radon-222 concentration 191 Bq m <sup>-3</sup> Maximum outside radon-222 concentration 48 Bq m <sup>-3</sup>	46
Canada (9999 dwellings)	64% of dwellings support radon-222 concentrations less than 37 Bq m <sup>-3</sup> 95% of dwellings support less than 0.1 WL Highest radon-222 concentration recorded 2780 Bq m <sup>-3</sup> Highest WL value measured 0.233 WL	44
Germany (No. of measurements not stated)	Measurement of gross radon-222 decay products concentration in the range 0.1 - 10 WL	47
Hungary (849 dwellings)	92% of dwellings had been shut up for at least 8 hours before measurements were carried out. Mean concentration of polonium-218 113 Bq m <sup>-3</sup> Mean concentration of lead-214 98 Bq m <sup>-3</sup> Mean concentration of bismuth-214 92 Bq m <sup>-3</sup> Maximum concentration of all nuclides >370 Bq m <sup>-3</sup> The distribution of the measured values was markedly skew with the majority of the results less than 37 Bq m <sup>-3</sup>	50
Norway (120 dwellings)	Wood construction Mean radon-222 concentration 48 Bq m <sup>-3</sup> range 7.4 - 137 Bq m <sup>-3</sup> Concrete construction Mean radon-222 concentrations 74 Bq m <sup>-3</sup> range 7.4 - 248 Bq m <sup>-3</sup> Brick construction Mean radon-222 concentration 37 Bq m <sup>-3</sup> range 11 - 215 Bq m <sup>-3</sup>	49
Poland (No. of measurements not stated)	Radon-222 concentrations in buildings in the range 3.0 - 79 Bq m <sup>-3</sup> Measured radon-222 production rates in dwellings indicate that on average a member of the Polish population receives an annual exposure to environmental radon-222 decay products in the range 0.12 - 0.21 WLM	48 52
USA Tennessee	Mean WL found in dwellings 0.0071 WL	51
Schenectedy	Mean radon-222 concentration in ground- floor rooms (3 rooms) 28.5 ± 3.3 Bq m <sup>-3</sup> In basements (2 rooms) 54.0 ± 4.4 Bq m <sup>-3</sup>	43

## 5.2 Measurements reported in the UK

It was shortly after the work of Hultqvist in Sweden that investigators in other countries, including the UK, carried out programmes of measurement of the radon-222 concentration in room air and in outside air. The early measurements were limited to small numbers and restricted geographical locations. Haque et al<sup>(53)</sup> investigated rooms in south London. In outside air the average concentration was  $3.3 \text{ Bq m}^{-3}$ , with minimum and maximum values of 0.37 and  $11 \text{ Bq m}^{-3}$ , respectively. Within rooms the radon-222 concentration was always higher than that in the outside air. Both indoor and outdoor radon-222 concentrations showed wide excursions and this pattern was observed in both well-ventilated and poorly ventilated rooms. The rise in radon-222 concentration in some sealed rooms was monitored until a steady concentration was obtained after 8 days. The calculated mean radon exhalation rate per unit surface area of these rooms were in the range  $5.86 \cdot 10^{-5}$  to  $2.36 \cdot 10^{-4} \text{ Bq m}^{-2} \text{ s}^{-1}$ . Obviously not all room surfaces were of the same material, but a mixture of clay bricks and concrete would be unlikely to produce the reported emanation rates (see Table 10). It is much more likely that the flux from the building materials was augmented by radon migrating from the subsoil through the floor into the room.

Other measurements of radon-222 concentration in air within a small number of dwellings and in outside air were made in the 1960s. Several hundred such measurements were analysed and these demonstrated that concentration excursions exceeding an order of magnitude were common<sup>(54)</sup>. These results indicated that the analysis of a single air sample taken at a dwelling would offer no indication of the average radon-222 concentration at that sample site. The rate of production of radon-222 within a room containing given materials will be influenced by meteorological variables, such as atmospheric pressure, but will be subject to much smaller variation than the day-to-day radon-222 concentration in room air. A pilot study was inaugurated by Duggan and Bradford<sup>(55)</sup> in which measurements were carried out to assess the rate of production of radon-222 in room air in terms of activity concentration per unit volume of room air per unit time. As the radon-222 production rate is not constant (but probably varies by no more than a factor of three), a series of measurements in a number of dwellings should produce a realistic overall mean radon-222 production rate from which an estimate of the population exposure to the decay products of radon-222 can be derived, although individual exposures in a particular dwelling cannot be estimated precisely.

Following the pilot study, a wider programme involving measurements in some 80 dwellings was undertaken<sup>(56)</sup>. The aim of this survey was to determine the average population exposure to the decay products of radon-222. With such a small sample, from a total housing stock of approximately 20 million, the survey was constrained mainly to dwellings in the major centres of population, including London, Birmingham, Liverpool, Manchester, Glasgow, Edinburgh and Aberdeen, and a

limited number of rural sites. Most of these areas are sedimentary and the majority of the population also live in sedimentary areas. The mean radon-222 production rate was then calculated to be  $20 \text{ Bq m}^{-3} \text{ h}^{-1}$ , after excluding one particularly high value found in a remote region. A number of assumptions have to be introduced to enable an average exposure to the decay products of radon-222 to be derived from the production rate. The assumptions are that the average ventilation rate in UK dwellings is one air change per hour, the average occupancy factor for UK dwellings is 90%, and that radon-222 decay products are lost from room air predominantly by radioactive decay and ventilation, with removal by plate-out to room surfaces playing a minor role. This resulted in the prediction of an average annual exposure of the UK population to the decay products of radon-222 of 0.16 WLM, or an annual effective dose equivalent of 800  $\mu\text{Sv}$ . However, as pointed out earlier, exposure to the decay products of radon-222 exhibits a much wider spread in values than exposures to other sources of natural radiation. The range of production rates found in this national survey was  $0.44\text{--}104 \text{ Bq m}^{-3} \text{ h}^{-1}$ , giving a range of annual effective dose equivalents of 13  $\mu\text{Sv}$  to 4.1 mSv. The annual potential exposure in the one dwelling excluded for the purpose of deriving the average population exposure being 8.1 mSv.

Calculation of the potential exposure to radon-222 decay products within buildings cannot be used to determine individual exposures with any degree of confidence. Individual preferences for ventilation and time spent in the open air will drastically affect each individual's exposure, but the average exposure of 0.16 WLM  $\text{y}^{-1}$  (or 800  $\mu\text{Sv y}^{-1}$ ) for members of the UK population is similar to values determined for populations in other developed countries. There would seem to be little cause for concern over exposures to radon-222 decay products that are close to the average value or below it. Some exposures at the high end of the distribution deserve further consideration as here the exposure may be deemed to be detrimental.

In the survey work reported above most measurements of potential radon-222 decay products exposure were carried out in sedimentary areas where the underlying soil and rock is not noted for extensive uranium mineralisation. Even here it can be seen that the influence on the radon-222 concentration in room air of radon-222 from the ground beneath the dwelling can be marked. Thus, in those areas where uranium mineralisation is extensive, much higher radon-222 concentrations in dwellings and other buildings might be expected. In many of these areas the population will be low and high radon-222 concentrations here will not dramatically affect the estimate of the average population exposure.

Meetings have been held with members of staff of the Institute of Geological Sciences who have studied the available geological data and suggest that certain areas might have an elevated radon-222 flux from the ground, and hence contribute to enhanced radon and radon decay product concentrations in indoor air. These are:



Aberdeenshire — including Aberdeen City, but more likely in the Banchory area;

Kirkcudbrightshire;

Pennines — Carboniferous limestone and Derbyshire Domes;

Orkney;

Wales — south Wales in the area of Swansea, certain areas in north and mid-Wales;

Cornwall — certain areas of the county;

Devon — Dartmoor Aureole;

Caithness.

Measurements carried out in some of these areas are detailed in Section 5.2.1. below.

#### 5.2.1 Measurement of the concentrations of radon-222 and its decay products in air in regions of extensive uranium mineralisation

The radon-222 production rate of a room is determined from measurements of the polonium-218 concentration in the room air and in outside air, together with a measurement of the air exchange rate of the room. The air exchange rate is determined by the standard tracer gas technique<sup>(57)</sup> and the polonium-218 concentration by air sampling through a filter paper. The radon-222 production rate is affected by meteorological variables (notably pressure) but seldom varies by more than a factor of three about the mean. This is a much smaller variation than that obtained from isolated determination of the radon-222 gas concentration in room air.

In areas associated with uranium mineralisation, 462 dwellings have been visited and measurements carried out to determine radon-222 production rates. As was expected, the production rates found in these areas are generally above those found in the national survey, and hence lead to higher concomitant potential exposures to radon-222 decay products. For comparison purposes, the cumulative distribution of potential annual effective dose equivalents from the survey in these limited regions is shown in Figure 1, together with the distribution from the national survey. Exposures have been given as effective dose equivalent, using the conversion factor of 1 WLM = 5 mSv, so that direct comparison with other sources of natural radiation exposure may be effected. It should be noted that the mean annual effective dose equivalent in the areas of uranium mineralisation is 6.4 mSv compared with 0.8 mSv in the national survey.

As has been stressed, the exposures calculated above are subject to large uncertainties as the actual exposure of the occupier of a house is critically dependent upon his or her preference for ventilation. In order to investigate the agreement between the exposure calculated from a single visit to a house with air sampling equipment and the true mean radon-222 gas concentration, passive radon-222 monitors were left in a number of dwellings. In most cases a detector was left in the main bedroom and another in the main living room, with

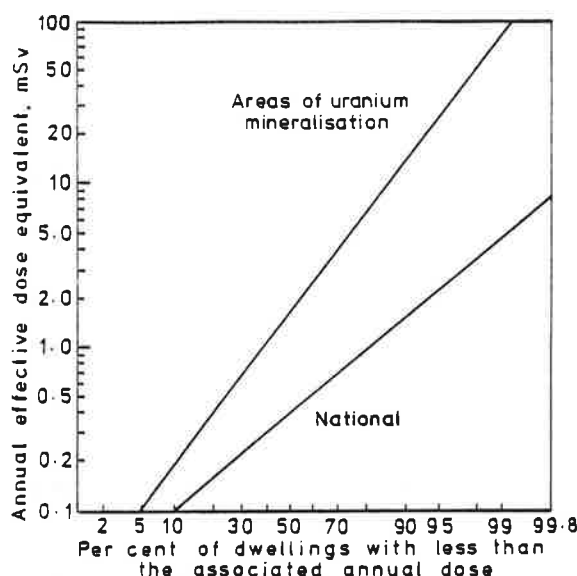


Figure 1 Cumulative distributions of annual effective dose equivalent from  $^{222}\text{Rn}$  decay products exposure

further detectors occasionally left in other rooms. The radon-222 concentration in room air is expected to show a marked seasonal variation as windows are more frequently opened in the summer than the winter. Initially, detectors were installed for three months and then replaced with unexposed detectors which were in place for the next three months, and so on until a year had been covered. Later, the period of monitoring by each detector set was extended to six months. The detectors used are described in Appendix B, Section 4.

Following the initial survey of dwellings in areas of uranium mineralisation, two groups of dwellings were chosen for additional study. These were dwellings where the active measurements (by air sampling) indicated potential radon-222 decay products exposures exceeding 25 mSv in a year and those dwellings with a potential annual exposure between 5 mSv and 25 mSv. Table 16 displays the results of the active and passive measurements for the former case and Table 17 presents the results for the latter case.

In Tables 16 and 17 the active measurements given in column eight were invariably carried out in the living room and, therefore, these predicted results should be compared with the results from the passive detectors exposed in the lounge. It must also be noted that exposure periods 1, 2, etc do not imply the same three-month calendar period for all dwellings. Some dwellings will have started period one in May, others in October, others in January, and so on. However, periods 1, 2, 3 and 4 referring to one dwelling are contiguous.

Table 16

Results from passive radon-222 detectors exposed for three-monthly periods in dwellings 1-9 where active measurements indicated a potential annual effective dose equivalent (EDE) in excess of 25 mSv from exposure to the decay products of radon-222

	Room	Potential EDE received during the period (mSv)				Total EDE (12 months)	Potential annual EDE indicated by active measurement (mSv)
		1	2	3	4		
1	L	11.6	11.2	15.5	9.80	48.1	28.5
	BE	3.82	3.72	3.63	5.54	11.7	
2	L	89.5	106	56.2	N/A	> 252	(i) 45.1
	BE	98.0	111	18.5	82.0	310	(ii) 364
	K	90.5	34.3	33.7	87.5	246	
	BA	61.3	35.2	18.5	13.9	129	
3	L	2.8	5.01	7.7	2.93	18.4	36.1
	BE	1.45	1.85	2.16	0.69	6.15	
4	Study	16.0	2.46	N/A	10.6	> 29.1	160
	BE	9.55	1.69	15.9	3.88	31.0	
5	L	30.0	33.8	22.9	5.65	92.4	122
	BE	9.2	7.98	12.5	6.76	36.4	
6	L	12.2	8.11	0.58	3.11	24	(i) 29.3 (ii) 34.3
7	L	4.30	3.22	2.51	N/A	> 10.0	57.4
	BE	1.20	1.00	1.21	N/A	> 3.41	
8	L	16.2	9.6	2.83	1.94	30.6	88.5
9	L	16.3	6.2	9.01	N/A	> 31.5	57.9
	BE	3.14	1.96	4.09	N/A	> 9.19	

N/A = not available; K = kitchen; L = living room; BA = bathroom; BE = bedroom;  
(i) = first measurement and (ii) = subsequent measurement on a different day.

The passive radon detectors respond solely to the radon-222 gas concentration in room air, and the only strictly valid interpretation of the results from these detectors is the average radon-222 gas concentration during the period of exposure. Ideally the parameter required is the average WL value for the period of exposure, but no passive device for recording this has so far been devised. It is therefore necessary to apply a scaling factor to the average radon-222 gas concentration to arrive at a reasonable estimate of mean WL value. To this end it has been assumed that the ventilation rate is one air change per hour and that the loss of radon-222 decay products from room air by plate-out to room surfaces is negligible. Under these conditions the WL is given by

$$WL = \frac{0.54 R}{3700} \dots\dots\dots(1)$$

where R is the radon-222 concentration in Bq m<sup>-3</sup>.

Table 17

Results from passive radon-222 detectors exposed for three-monthly periods in dwellings 10-18, where the active measurements indicated a potential annual effective dose equivalent (EDE) between 5 mSv and 25 mSv from exposure to the decay products of radon-222

	Room	Potential EDE received during the period (mSv)				Total EDE (12 months)	Potential annual EDE indicated by active measurement (mSv)
		1	2	3	4		
10	L	2.36	1.25	13.9	1.81	19.32	5.95
	BE	0.127	0.183	0.72	0.88	1.91	
11	L	6.70	1.22	1.52	1.05	10.5	(i) 6.73
	BE(1)	3.65	6.21	0.37	2.14	12.4	(ii) 5.33
	BE(2)	0.90	1.60	0.58	3.80	6.88	
	K	1.4	1.35	0.13	2.47	5.35	
12	L	4.4	33.7	2.01	2.10	42.2	(i) 15.0
	BE	1.06	0.705	0.45	0.87	3.09	(ii) 9.02
13	L	1.27	0.88	0.84	1.48	4.47	6.62
	BE	0.70	0.34	1.04	0.89	2.97	
14	L	1.90	1.57	0.52	0.80	4.79	5.58
	BE	3.10	3.14	1.04	1.25	8.53	
15	L	0.10	7.01	10.2	3.92	21.2	6.06
16	L	1.50	11.8	0.985	10.3	24.6	
	BE	4.24	3.31	0.765	1.84	10.2	18.9
	BA	6.21	1.01	1.78	1.86	10.9	
17	L	28.1	2.77	2.41	7.50	40.8	9.02
	BE	3.10	1.65	1.13	6.28	12.2	
	D	2.64	6.02	N/A	8.23	> 16.9	
18	L	0.565	0.019	2.12	1.13	3.83	6.30
	BE	0.195	0.365	0.35	0.33	1.24	

N/A = not available; K = kitchen; L = living room; BA = bathroom; BE = bedroom; D = dining room; (i) = first measurement and (ii) = subsequent measurement on a different day.

In practice there will be a range of ventilation rates, and removal of decay products by plate-out may be a significant mechanism in some circumstances. The factor 0.54 is increased for decreasing ventilation rate and decreased for increasing plate-out. In Tables 16 and 17 the potential exposures assume 90% occupancy.

Active measurements were carried out on more than one occasion at some dwellings, as indicated in column eight of the tables. As outlined earlier, the calculated value for the radon-222 production rate, based on a series of single half-day measurement periods, has been found in the past to produce similar results within a factor of three. This pattern is seen in dwellings 6, 11 and 12, but for dwelling 2 there is a factor of eight between the two measurements.

No totally satisfactory explanation for this large discrepancy has been found, except that on the first visit there was noticeable plate-out of radon-222 decay products on the clean filter papers. This suggests that a large proportion of the airborne decay products were being lost to room surfaces, thus reducing the concentration of these nuclides detectable by air sampling. Inspection of the passive detector results for dwelling 2 show that the predicted potential exposure of 364 mSv is more realistic than the initial estimate of 45.1 mSv. The entries in Tables 16 and 17 marked N/A represent a failure in the processing of the detector. Each detector package contains two detector elements, only one of which is initially processed. The detector element first processed is subjected to a treatment that produces enhanced sensitivity, but also results in the catastrophic failure of a proportion of the detectors. When a failure occurs, the second detector element is processed using a method that is more time consuming but is unlikely to cause failure. Results from the processing of the second detector elements are still awaited.

It is apparent that the likely exposure of the occupants of dwellings to radon-222 decay products cannot readily be assessed from active measurements made over a short period of time. The active measurements do provide a useful screening procedure, in that in only one case where the active measurement indicated a likely exposure in excess of 5 mSv did the passive detector results indicate an exposure below 5 mSv. In this case (dwelling 18) the lower result was due to an abnormally low measurement in one quarter of 0.019 mSv. In all cases displayed in Tables 16 and 17 the large variation from quarter to quarter can be seen to reflect window opening behaviour under different climatic conditions. It is also apparent that exposures during the same period can be quite different according to the room occupied. In most cases the exposure in the bedroom is lower than that in the living room (with the notable exception of dwelling 14), which may reflect the lower radon-222 concentration expected on the first floor, due to it being more remote from the ground, but is more likely due to the common habit of opening windows in unoccupied bedrooms during the day regardless of the climate.

It is apparent from Tables 16 and 17 that in a number of dwellings an annual effective dose equivalent exceeding 50 mSv could be received by the occupants. The figure of 50 mSv corresponds to the maximum annual effective dose equivalent permitted to workers exposed to ionizing radiation. The situation in housing is aggravated by the longer periods of occupancy in houses than in the work place.

#### 5.2.2 Measurements of radon-222 decay products in high-rise buildings and in offices

It has been stated many times that the major source of radon-222 in buildings is the subsoil beneath the structure. It might be concluded from this that in high rise buildings the radon-222 concentration would decrease as one moved to floors of higher elevation and this was found to be the case by

Toth<sup>(50)</sup>. However, other work has indicated that this cannot be regarded as the general case. Measurements have been carried out in university residences at the University of Birmingham<sup>(58)</sup>. These residences have the advantage that on each floor above ground level the rooms are of similar layout and identical dimensions. The results of one series of measurements are given in Table 18.

Table 18  
Radon-222 decay products concentration measured in similar rooms  
on different floors of High Hall, Birmingham University

Day	Floor No.	Radon daughter activity (mWL)	Ventilation rate measured	Radon daughter activity normalised to a ventilation rate = $1.0 \text{ h}^{-1}$ (mWL)
2	Basement	15.1	0.23	2.2
1	1	0.82	0.79	0.58
1	2	1.02	0.58	0.47
1	3	1.74	-	-
1	4	2.73	0.43	0.85
1	5	3.19	0.40	0.92
2	6	7.71	0.41	2.3
2	8	3.98	0.36	1.0
2	9	4.04	0.21	0.53
2	10	2.08	0.58	0.96
2	12	3.31	0.40	0.95
2	14	5.64	0.65	3.1

Ideally, simultaneous measurements should have been made in all rooms on the same day, but limitations on equipment necessarily precluded this. It can be seen from Table 18 that there is no correlation between radon-222 decay products concentration and floor elevation, although the expected high concentration of radon-222 decay products in the basement is apparent. In column five the radon-222 decay product concentration for each room has been corrected to the value that would obtain at a ventilation rate of one air change per hour. This normalisation is only a first approximation as no measurements of the WL value in outside air were made, but the error is unlikely to be greater than 10%. Under normalised conditions the WL value in the room on the 14th floor exceeds that in the basement. The main reason that the radon-222 concentration does not decrease with elevation is that each floor is linked by stair-wells and lift shafts. Also, the rooms in this series of measurements had common services which were brought up from the ground floor and encased in each room by ill-fitting shuttering. Thus, there were many routes by which radon-222 could migrate from one floor to another.

A second series of measurements were made in halls of residences at Birmingham University choosing sides of the building where there were no common service ducting that could provide a route for radon-222 between floors. In these measurements the WL value found in floors of different elevation produced erratic values which did not support the hypothesis that the level would be systematically lower on higher floors.

In halls of residences the stair-wells are internal to the building and access to rooms is via internal corridors. Under these circumstances it is doubtful that residents on the upper floors receive a significantly lower radon-222 decay product exposure than those on the lower floors. In blocks of flats with open stair-wells and where access to the flat is along an open balcony the radon-222 concentration on higher floors may be lower than that on floors near the ground. Even here, common services are brought to each floor. If this is achieved by ducting linking floors of different elevation, again there will be an easy route by which radon-222 from the ground can reach the higher housing units.

Most emphasis in measurements of radon-222 and its decay products has centred on concentrations found in dwellings. However, for many people a substantial proportion of their time is spent away from home in buildings associated with their place of work. A large number of people work in small offices which have a larger surface area to volume ratio than rooms in domestic premises. In these circumstances the contribution of the building materials to the radon-222 concentration in an office will be larger than that in a living room. Office doors are often closed for prolonged periods to reduce noise levels from without, and air exchange rates in offices may be low. The Director General of the Greater London Council gave permission for the carrying out of measurements in offices within the County Hall complex. County Hall has the advantage of being a multi-storey building that has been added to on a number of occasions during its history, such that different parts of the building are constructed from different materials.

A total of 17 rooms in the County Hall complex were studied. In each office, measurements of the decay products of radon-222 were carried out and a measurement of the air exchange rate made. It was not possible to determine what proportion of the air exchange was between the room and the corridor rather than with the outside air. Measurements of radon-222 decay products in a few corridors indicated that the concentrations here were not significantly different from those in the outside air. However, in some offices air exchange between adjacent offices could not be ruled out. A description of the rooms studied is given below and the measurement results summarised in Table 19.

Room 1. Single-person office on the third floor of the main building having double glazed windows in the exterior wall facing northeast. Volume — approximately 32 m<sup>3</sup>.

Room 2. Single-person office on the third floor of the main building having double glazed windows in the exterior wall facing west. Volume — approximately 34 m<sup>3</sup>.

Room 3. A large laboratory on the seventh floor of the main building. These rooms are a recent addition to the building and are predominantly glass and wood, except where attached to the main building at roof height. Volume — approximately 260 m<sup>3</sup>. Two single-person offices were partitioned off the laboratory, but the connecting doors between the offices and the laboratory were open throughout the measurement period.

Room 4. A large office, partitioned off a laboratory similar to room 3. Window in exterior wall facing northeast had a 5 cm gap at the top throughout the measurement period. Door to the laboratory was closed. Ceiling is covered in acoustic tiles, wall partitions are of sheet metal and the floor is concrete. Volume — approximately 68 m<sup>3</sup>.

Room 5. Single-person office on the sixth floor of the main building with exterior wall facing southwest. Volume — approximately 63 m<sup>3</sup>.

Room 6. Single-person office on sixth floor of main building with exterior wall facing southwest. Volume — approximately 42 m<sup>3</sup>. The floor covering is wood block.

Room 7. Basement room in South Block with concrete floor, exposed brick walls and a skylight in the ceiling at road height.

Room 8. A conference room on the ground floor of the main building. The exterior wall faces north and has two extractor fans either side of a closed window. Neither fan was operated during the measurements nor for several hours beforehand. Volume — approximately 200 m<sup>3</sup>.

Room 9. An office on the fourth floor of the main building with exterior wall facing north. The double glazed window was slightly open, with open areas approximately 0.1 m<sup>2</sup>. Volume — approximately 85 m<sup>3</sup>.

Room 10. An office on the ground floor of North Block with exterior wall facing east. Volume — approximately 60 m<sup>3</sup>.

Room 11. An office on the first floor of North Block with exterior wall facing east. Volume — approximately 53 m<sup>3</sup>.

Rooms 12 and 13. These are identical rooms on the fourth and third floors, respectively, of the Island Block. This is the most recently added block to the complex and has a full integral air conditioning plant. This plant was originally designed to control temperature and humidity and includes a washing plant. The washing plant is no longer in use because of prohibitive running costs. Air is now simply circulated through filter media. Minimum make-up air is 10%, but outside air is used to reduce or increase temperature as required. Thus maximum make-up air may approach 100%. The summer design temperature is 23°C, but a range of 20–24°C is typical. The volume of each room is 40 m<sup>3</sup>. The high air exchange rates from the air conditioning plant result in very low



concentrations of the decay products, and the uncertainties on the determination of the individual nuclide concentrations approaches or exceeds the measured values. Thus, results of the mWL value determinations are given for these rooms only, as here the uncertainties are better than 17%. The difference in ventilation rates between the two rooms is real and not an artifact of the measurement, although the reason for the difference was not deduced. The calculated values of the ventilation rates together with the standard deviations were  $13.0 \pm 0.77 \text{ h}^{-1}$  and  $6.48 \pm 0.44 \text{ h}^{-1}$  for rooms 12 and 13, respectively. Although the ventilation rates differ by a factor of two, the mWL values are similar and could not be distinguished from the value in the open air.

Room 14. A first floor room in South Block with exterior wall facing east. Volume — approximately  $55 \text{ m}^3$ .

Room 15. A first floor room in South Block with exterior wall facing west. This office was across the corridor from room 14 and of similar volume — approximately  $55 \text{ m}^3$ .

Room 16. A basement room in South Block used as a mess room. Volume — approximately  $120 \text{ m}^3$ . Although classed as a basement because the rooms are below street level, there is a walk-way around the outside of the basement and all rooms have windows. No room is completely surrounded by soil.

Room 17. Large room just off the main entrance hall of the main building with exterior wall facing east. Volume — approximately  $240 \text{ m}^3$ .

Table 19  
Radon-222 decay product concentrations in rooms of County Hall

Room	Floor	Mean activity concentration ( $\text{Bq m}^{-3}$ )			mWL	Air exchange rate ( $\text{h}^{-1}$ )
		Polonium-218	Lead-214	Bismuth-214		
1	3	6.10	4.58	3.77	1.18	0.52
2	3	4.35	3.79	1.78	0.824	0.76
3	7	11.0	7.75	3.58	1.91	0.73
4	7	4.30	3.03	2.58	0.904	3.42
5	6	9.09	8.20	6.76	2.03	0.47
6	6	6.83	5.66	2.26	1.02	0.39
7	SB	5.12	4.48	3.65	1.30	2.48
8	G	2.52	1.65	0.50	0.346	0.95
9	4	1.76	1.48	0.76	0.329	1.23
10	G	4.29	3.72	2.75	0.908	1.09
11	1	6.29	7.11	3.64	1.52	0.39
12	4	-	-	-	0.096	13.0
13	3	-	-	-	0.103	6.48
14	1	2.48	2.14	1.64	0.529	0.91
15	1	2.39	1.45	2.34	0.502	0.64
16	B	0.93	1.10	0.917	0.270	3.72
17	G	2.40	1.91	1.74	0.452	1.07

G = ground floor; B = basement; SB = sub-basement.

It will be noted in Table 19 that for some rooms the activity concentration measured is less for polonium-218 than lead-214, although this condition cannot exist on theoretical grounds. These anomolous results arise due to counting statistics when measuring low concentrations subject to large uncertainties. The mWL level is always measured with an acceptable uncertainty. These points are illustrated in Table 20 where the full results of the measurements in room 16 of County Hall are presented.

Table 20  
Full measurement results for room 16 of County Hall

Measurement	Activity concentration (Bq m <sup>-3</sup> )			mWL
	Polonium-218	Lead-214	Bismuth-214	
1	1.14±0.86	1.04±0.302	0.851±0.907	0.260±0.025
2	0.392±0.884	1.25±0.323	1.04 ±0.429	0.288±0.026
3	1.27±0.858	1.01±0.298	0.858±0.400	0.261±0.024

A measurement of the radon-222 decay product concentrations in outside air was obtained by placing the monitor on the window ledge of room 15 with the sampling head well clear of the building. The results were:

Polonium-218    0.429 ± 0.951 Bq m<sup>-3</sup>  
Lead-214        1.095 ± 0.332 Bq m<sup>-3</sup>  
Bismuth-214    1.413 ± 0.451 Bq m<sup>-3</sup>  
mWL             0.305 ± 0.021.

Occasional measurements at ground level produced results similar to these. Thus in many rooms within the County Hall complex the concentration of the decay products of radon-222 were little different from those in the open air. Here again there is no systematic variation in radon-222 decay products concentrations with floor elevation, although such a pattern, if it exists, may be masked as consequence of measurements taking place over a two-week period.

The average mWL for all rooms measured in the County Hall complex is 0.84, which is lower than that found in domestic premises. For a 2000 h working year this would indicate a typical annual exposure of about 0.01 WLM, or 50 µSv effective dose equivalent. Other office blocks would undoubtedly produce different results. However, County Hall is constructed of a variety of materials; the outer shell being granite on the side facing the river and sandstone on the other side (facing York Road), with concrete of different ages throughout the building. The Island Block is predominantly steel and glass. Thus it is doubtful that a more diverse structure exists, and from the results here there is no reason to suggest that occupancy of offices leads to a significant increment in radon decay product exposure.

### 5.2.3 The concentration of the decay products of radon-220 in the general environment

As was stated earlier, thorium-232 is also a trace contaminant of most building materials and a member of its decay chain is the isotope of radon, radon-220. Radon-220 has a half-life of 54.5 s and only those atoms formed close to the surface of a material can escape to the surrounding air. However, the number of atoms of a radionuclide required to form unit activity (1 Bq) is directly proportional to the half-life. This means that should a surface be emitting equal flux densities of atoms of radon-222 and radon-220 the activity flux density of radon-220 is approximately 6000 times that of radon-222. In practice, fewer atoms per unit time of radon-220 will be emitted from a surface than atoms of radon-222, but it is possible for the emanation rate (in  $\text{Bq m}^{-2} \text{s}^{-1}$ ) of each nuclide to be of the same order. For this reason it has been suggested that the decay products of radon-220 may pose a significant source of exposure to the occupants of buildings<sup>(59)</sup>. Having such a short half-life the flux of radon-220 into room air is drastically reduced by any lining material, such as wallpaper. Stranden<sup>(59)</sup> has calculated the ratio of the WL due to radon-222 decay products to that due to those of radon-220 for a room with no lining materials, as a function of ventilation rate. This relationship is shown in Figure 2. The reason why the ratio increases with increasing ventilation rate

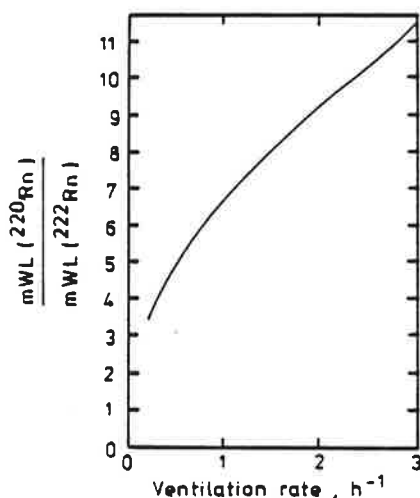


Figure 2 Theoretical WL ratio in dwellings as a function of ventilation rate

is that the concentration of the decay products of radon-222 falls more rapidly as the ventilation rate increases than does the concentration of the decay products of radon-220. One further difference between the decay products of radon-222 and those of radon-220 is that 3763 Bq m<sup>-3</sup> each of polonium-218, lead-214 and bismuth-214 form 1 WL for radon-222 decay products. For the decay products of radon-220, 284 Bq m<sup>-3</sup> each of lead-212 and bismuth-212 form 1 WL. The derivation of the WL is given in detail in Appendix A. Instruments have been acquired that permit the assessment of the concentrations of the decay products of radon-222 and radon-220 from the same air sample, and hence ratios of the WL values attributed to each series can be obtained. Table 21 lists results obtained in a number of dwellings throughout England. The traditional names for radon-220 and radon-222 are thoron and radon, respectively, and in Table 21 mWL-Tn refers to the mWL arising from the decay products of radon-220 and mWL-Rn to the mWL arising from the decay products of radon-222.

From Table 21 it is seen that in no case did the ratio of the working levels approach the large values predicted for rooms with uncovered walls. There is a discernible trend for the larger ratios to occur at higher ventilation rates, but the trend is weak and there are notable exceptions. This is to be expected when measurements are carried out in different dwellings, each of which will have unique source terms for radon-222 and radon-220 production rates within the building. All measurements of the concentration of radionuclides are subject to uncertainties arising from the random nature of radioactive decay. Uncertainties have not been shown in Table 21 in order to make the table less confusing. To convey an idea of the uncertainties in these figures a few complete examples are given in Table 22.

It can be seen from Table 22 that some care is needed in interpreting results where ratios are derived from data which are subject to a high degree of uncertainty. The unweighted mean of the working level ratios given in Table 21 is 1.00 (SD = 0.84). Further measurements are required under a wider variety of conditions to confirm that this figure is truly representative of conditions in British dwellings. If it is, then this would indicate that the currently accepted value for the annual average effective dose equivalent received by a member of the UK population should be increased from 800 µSv (due to radon-222 alone) to 1070 µSv, taking due note that the risk per unit exposure (in WLM) to the decay products of radon-220 is one third that from an equal exposure to the decay products of radon-222. These figures are of course tentative, but the evidence is firm that the decay products of radon-220 do form a source of human exposure to natural radiation that is non-trivial.

Table 21  
Radon and thoron decay products working levels calculated  
from the same air sample

mWL-Tn	mWL-Rn	$\frac{\text{mWL-Tn}}{\text{mWL-Rn}}$	Ventilation rate ( $\text{h}^{-1}$ )
1.75	1.34	1.31	1.98
3.88	1.83	2.12	1.24
0.29	0.21	1.38	1.38
3.18	1.68	1.89	0.98
2.28	4.10	0.56	2.76
0.66	2.00	0.33	0.47
16.0	17.1	0.93	0.12
0.18	0.50	0.36	5.65
1.41	2.52	0.56	0.82
0.98	3.40	0.29	0.12
0.85	1.67	0.51	5.58
1.14	5.06	0.23	0.80
0.31	0.33	0.94	0.77
0.23	0.61	0.38	0.70
1.19	1.25	0.95	0.77
0.17	0.79	0.22	1.14
1.38	0.49	2.83	3.98
0.19	0.44	0.43	1.61
1.71	0.96	1.78	10.3
0.12	0.17	0.67	1.59
2.56	3.79	0.68	2.76
2.62	14.8	0.18	0.89
0.50	1.70	0.31	0.41
0.50	1.82	0.27	2.16
0.14	0.29	1.19	0.48
4.47	45.8	0.10	0.40
1.51	0.52	2.92	2.41
1.66	2.05	0.81	0.88
3.59	2.03	1.77	1.51
1.20	2.61	0.46	0.64
0.173	0.056	3.04	1.53
0.64	0.31	2.02	1.92
1.13	0.38	2.96	0.47
0.58	2.66	0.58	0.22
0.72 <sup>1</sup>	0.82	0.88	0.76
1.29 <sup>1</sup>	1.91	0.68	0.73
0.41 <sup>1</sup>	0.90	0.46	3.42
2.15 <sup>1</sup>	2.03	1.06	0.47
0.54 <sup>1</sup>	1.02	0.53	0.39
0.71 <sup>1</sup>	1.30	0.55	2.48

<sup>1</sup>These measurements were made in offices of the County Hall complex. The remainder were in dwellings.

Table 22

A selection of radon and thoron decay products measurements  
together with the associated statistical uncertainties

mWL-Tn	mWL-Rn	$\frac{\text{mWL-Tn}}{\text{mWL-Rn}}$
$1.51 \pm 0.131$	$0.518 \pm 0.193$	$2.9 \pm 1.09$
$3.59 \pm 0.203$	$2.03 \pm 0.299$	$1.77 \pm 0.27$
$0.173 \pm 0.0421$	$0.056 \pm 0.061$	$3.04 \pm 3.30$
$0.575 \pm 0.086$	$2.66 \pm 0.098$	$0.216 \pm 0.032$
$1.20 \pm 0.120$	$2.61 \pm 0.178$	$0.458 \pm 0.055$

5.2.4 Studies of radon decay product concentrations in houses under 'whole-house' conditions

In surveys of radon decay product concentrations in dwellings occupied by members of the general public, measurements have been confined to one room. This was necessary in order to cause the minimum inconvenience to the occupiers, who volunteered for studies from which they received no benefit. In studying a single room, it was assumed that the air exchange was between that room and the outside, although obviously some air exchanges with adjoining rooms even when the room chosen is on the windward side of the building. Where passive radon detectors have been installed in single rooms that have been studied previously with active devices the agreement between the predicted concentration of radon-222 and those actually found by the detectors have generally been good. Nevertheless it was considered that the radon-222 production rate of the entire dwelling could be determined if measurements were carried out in dwellings where all internal doors were open, and external doors and windows were closed. This was termed the 'whole-house' condition, and ventilation rates measured under these conditions, by releasing a tracer gas in all rooms and allowing mixing before monitoring the concentration, must represent air exchange solely with the outside.

Twenty members of Board staff volunteered the use of their dwellings and measurements were carried out under the above conditions with each measurement period occupying one day. At each dwelling radon-222 decay products concentrations were measured within the dwelling at two sites (on different elevations for multi-storey buildings) and in the open air. Ventilation rate was determined by the tracer gas technique, using nitrous oxide as the tracer and an infra-red gas analyser as the detector. A guide to wind conditions was noted using the Beaufort Scale as no recording anemometer was available. A description of the dwellings together with ventilation rate details are given in Table 23.

Table 23  
Description of dwellings 1-20 and ventilation details for those  
dwellings used for 'whole-house' studies

	Type	Whole-house ventilation rate ( $\text{h}^{-1}$ )	Features affecting ventilation rate	Reported wind force (Beaufort Scale)
1	Brick built dormer bungalow 2 bedrooms upstairs	0.161±0.013	Open fire in lounge	2+3
2	Brick built semi- detached	1.25±0.036	Double glazed	1
3	Metal and wood construction mobile home	0.536±0.0074	Air vents in wall and air vent above Parkray fire	1
4	Brick built detached	0.371±0.024	Downstairs front and bedrooms double glazed	1
5	Brick built terraced house	0.642±0.041	Part double glazed. Underfloor venti- lation in lounge	3+
6	Brick built semi- detached	0.962±0.045		1+4+3
7	Stone terraced cottage	2.41±0.020	Open fire (alight)	5
8	Brick built detached	0.883±0.021	Open fire in lounge (unlit). Bath- room only double glazed	4
9	Brick built end terrace	1.51±0.047	Window slightly open. Gas fire burning	3+2
10	Brick built detached	0.641±0.031	Open fireplace (unlit) in lounge	1
11	Brick built end terrace	0.383±0.018		1+2
12	2nd-floor flat	0.916±0.016	Part double glazed	1
13	Brick built detached bungalow	0.540±0.0083	Part double glazed Open fire place in lounge (unlit)	2
14	Brick built 3- storey terrace	0.990±0.030		5
15	Brick built detached bungalow	1.63±0.045	Suspended wooden floor. Open fire in lounge (unlit)	3+1
16	Brick built terraced	0.765±0.015	Double glazing throughout apart from front door	5+7
17	Concrete faced brick built semi-detached house	1.53±0.037	Open fire in lounge (unlit). Air vents in all rooms	5+6
18	Brick built end of terrace, 3- storeys + basement	1.91±0.064	Open fireplaces on second floor (unlit). Large high ceilinged rooms	5+6
19	Brick built detached bungalow	0.173±0.010	One window slightly opened. Part double glazed	2+4
20	Brick built semi- detached, 3 storeys + basement	0.465±0.020	Part double glazed	2

The usual procedure adopted for assessing the radon-222 production rate of a room, or in this case the entire dwelling, is to measure the polonium-218 concentration indoors and in the outside air, and to determine the ventilation rate. From these measurements the radon-222 production rate,  $K$  ( $\text{Bq m}^{-3} \text{ h}^{-1}$ ), is given by

$$K = j (1 + 0.0733j)(C_I - C_O) \quad \text{.....(2)*}$$

where  $j$  is the ventilation rate  $\text{h}^{-1}$

$C_I$  the concentration of polonium-218 in indoor air ( $\text{Bq m}^{-3}$ )

and  $C_O$  the concentration of polonium-218 in outdoor air ( $\text{Bq m}^{-3}$ ).

Equation (2) holds in the steady state. That is when conditions have been stable long enough for perturbations to the polonium-218 concentration to have settled down. The use of polonium-218 concentrations to determine the radon-222 production rate takes advantage of the short half-life of polonium-218. Under most conditions, where changes in ventilation rate are not too severe, the polonium-218 concentration will have sensibly reached the steady state two hours after the room conditions have been set. Unfortunately, the uncertainty with the measurement of polonium-218 concentration at low concentrations is often such that the standard deviation approaches the measured value. This of course leads to a large uncertainty in the value of  $K$ . From the calculated value of  $K$  the concentration of radon-222,  $R_j$  ( $\text{Bq m}^{-3}$ ), expected in indoor air at any practical ventilation rate,  $j$ , is given simply by

$$R_j = (K/J) + R_O \quad \text{.....(3)}$$

where  $R_O$  is the concentration of radon-222 in outside air, which can normally be taken as equal to the concentration of polonium-218 in outside air.

Where concentrations of polonium-218 in indoor air are so low that the uncertainties on any derived parameters are unacceptable an alternative approach can be used. At low concentrations of radon-222 decay products the mWL value in indoor air can be determined with an uncertainty better than 10% and that in outdoor air with an uncertainty better than 20%. However, for a change in ventilation rate conditions the indoor mWL value will take much longer to reach the steady-state value than will the polonium-218 concentration. Thus if mWL values are measured the random error due to counting statistics will be low, but there may be a large systematic error when the mWL value is used to predict for different conditions.

Nevertheless in this series of measurements both approaches ( $K$  and mWL) were used to predict the conditions that should prevail at a ventilation rate of one air change per hour.

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\*The derivation of all equations used in this report is given in Appendix B.



If  $mWL_I$  is the mWL value measured in indoor air at ventilation rate  $j \text{ h}^{-1}$  and  $mWL_O$  is the mWL value in outside air, the mWL value in indoor air at a ventilation rate of one air change per hour is given by  $mWL(1)$ , where

$$mWL(1) = mWL_O + [mWL_I - mWL_O] \quad \dots(4)$$

$$\times \left[ \frac{j (13.64 + j) (1.55 + j) (2.111 + j)}{2.670(1.551 + j)(2.111 + j) + 20.37(2.111 + j) + 31.64} \right]$$

Table 24 displays the results of these calculations for the dwellings in this study.

Table 24

The mWL values at one air change per hour predicted from the measured radon-222 production rate and the measured mWL values

Dwelling	Radon-222 production rate, K (Bq m <sup>-3</sup> h <sup>-1</sup> )	mWL at 1 ACH		Ratio K/mWL
		From K	From mWL(1)	
1	0.966 ± 0.448	0.572 ± 0.126	0.579 ± 0.009	0.988
2	3.06 ± 2.26	0.619 ± 0.367	0.716 ± 0.070	0.865
5	1.99 ± 1.25	0.615 ± 0.244	0.635 ± 0.053	0.969
6	16.3 ± 3.18	2.61 ± 0.512	2.07 ± 0.775	1.261
7	10.1 ± 4.33	1.52 ± 0.634	1.79 ± 0.077	0.849
8	9.36 ± 3.29	1.83 ± 0.551	2.19 ± 0.034	0.836
9	8.70 ± 5.55	1.97 ± 0.838	4.34 ± 0.117	0.454
10	10.7 ± 2.48	1.70 ± 0.421	1.84 ± 0.040	0.924
11	10.3 ± 1.65	1.85 ± 0.323	1.92 ± 0.018	0.964
12	3.51 ± 2.05	0.852 ± 0.368	0.861 ± 0.027	0.990
13	3.68 ± 1.12	0.661 ± 0.196	0.653 ± 0.010	1.01
14	11.7 ± 2.11	1.77 ± 0.321	1.54 ± 0.053	1.15
15	13.8 ± 3.04	2.15 ± 0.457	1.47 ± 0.120	1.46
16	1.35 ± 1.08	0.359 ± 0.191	0.338 ± 0.022	1.06
17	2.58 ± 1.78	0.424 ± 0.281	0.244 ± 0.093	1.74
18	9.69 ± 3.63	1.68 ± 0.541	2.14 ± 0.197	0.785
19	5.96 ± 0.921	1.09 ± 0.172	0.912 ± 0.007	1.120
20	1.19 ± 1.51	1.27 ± 0.392	2.07 ± 0.022	0.614

The agreement between the two approaches to calculate the mWL value that should exist at one air change per hour is remarkably good and does not solely apply to those dwellings where the measured ventilation rate was close to one air change per hour. For example, dwellings 12 and 13 both produce very close agreement, with the measured ventilation rate being close to unity (0.916) for 12 but  $0.54 \text{ h}^{-1}$  for 13. However, this series of measurements produced results that indicated much lower concentration of radon-222 decay products than had been expected on the basis of other measurements in dwellings in this area. To obtain an independent assessment of the average radon-222 concentration in rooms of these dwellings, passive radon-gas detectors were placed in rooms for a six-month period. When processed these detectors indicated average radon-222 concentrations generally in excess of the values predicted at the measured ventilation rate (V) by the active surveys, ranging from 0.96 to 20 times the predicted value. It was unfortunate that some of the passive radon-222 detector elements used here had a high background track density rendering the results subject to a larger uncertainty than is usual with these devices, but by no means large enough to entirely explain this discrepancy. The results for the 18 dwellings for which full results were obtained are given in Table 25.

The results of the active measurements under whole-house conditions were suspect in that the measured concentrations were lower than expected in the houses in this area, and lower than some measurements made in the same house when measurements were only carried out in one room. It appears that whole-house measurements produce a valid figure for the whole-house radon-222 production rate, but that this parameter is not a reliable indicator of the radon-222 concentration in individual rooms. A simple model, described in Appendix B, Section 5, demonstrates that single room measurements are more likely to produce realistic estimates of the radon-222 concentration in that room especially if it is on the ground floor. In the example given a whole-house radon-222 production rate measurement would predict a radon-222 concentration in the living room of  $17.3 \text{ Bq m}^{-3}$  at one air change per hour, a measurement of the radon-222 production rate in the living room would predict a concentration of  $22.4 \text{ Bq m}^{-3}$  whereas the true value is  $21 \text{ Bq m}^{-3}$ .

#### 5.2.5 A prolonged study in one dwelling

Studies were carried out in one dwelling over a period of 13 days, during which time it was unoccupied apart from by the investigators. Wind speed and direction charts for the period of this study were supplied by the Environmental and Medical Sciences Division of the Atomic Energy Research Establishment, Harwell. The meteorological station at which this data was collected is 8.5 km from the site of the house, but it is unlikely that the conditions at this spacing would be significantly different. The house is detached apart from a link garage to the neighbouring house. The north and south facing walls contain an extensive door and window area, there being very limited window area in the

Table 25

Average radon-222 concentration in dwellings predicted from active measurements and recorded by passive detectors  
(radon-222 concentration in outside air taken as  $1.8 \text{ Bq m}^{-3}$ )

Dwelling	Ventilation rate (V) at visit ( $\text{h}^{-1}$ )	Radon production rate, K ( $\text{Bq m}^{-3} \text{ h}^{-1}$ )	Mean radon concentration ( $\text{Bq m}^{-3}$ ) from K at ventilation rate		Mean radon-222 concentration from passive track detector ( $\text{Bq m}^{-3}$ )	Ventilation rate required, if K is correct, to give passive detector result
			$\text{V h}^{-1}$	$1.0 \text{ h}^{-1}$		
1	0.161	0.966	7.80	2.77	34	0.030
2	1.13	3.06	4.51	4.86	32	0.101
5	0.642	1.99	4.90	3.78	28	0.076
6	0.962	16.3	18.7	18.1	18	1.01
7	2.40	10.1	6.01	11.9	32	0.334
8	0.883	9.36	12.4	11.2	41	0.239
9	1.51	8.70	7.56	10.5	45	0.201
10	0.641	7.59	13.6	9.39	16	0.534
11	0.383	10.3	28.7	12.1	71	0.149
12	0.916	3.51	5.63	5.31	110	0.032
13	0.540	3.68	8.61	5.48	49	0.078
14	0.990	11.7	13.6	13.5	57	0.212
15	1.63	13.8	10.3	15.6	12	1.35
16	0.765	1.35	3.56	3.15	42	0.034
17	1.53	2.58	3.49	4.38	4	1.17
18	1.91	9.69	6.87	11.5	52	0.193
19	0.173	5.96	33.3	7.76	38	0.165
20	0.465	1.34	4.68	3.14	42	0.033

west and east facing walls. At the house, continuous records of humidity and barometric pressure were taken together with intermittent measurements of radon-222 decay products concentrations downstairs, upstairs and in outside air. A combined radon-222 and radon-220 decay products concentration measurements was performed late in the afternoon on most days. For all measurements all outside doors and windows were firmly closed and in the majority of cases all internal doors were opened to their fullest extent. The exception to this was for part of a day when the effect of an electrostatic precipitator on the decay products concentration in room air was investigated.

Figure 3 displays the recorded barometric pressure, wind speed and whole-house ventilation rate determined periodically during this period. The ventilation rate varied between 0.15 and 0.69  $\text{h}^{-1}$ , with a coefficient of variation on each measurement in the range 5–15%. The expected correlation between wind speed and ventilation rate is barely discernible, although the average ventilation rate is higher during days 2–5 when the wind was stronger than during the relatively calm period earlier and that of moderate winds from day 6 onwards. During the most windy period the ventilation rate was erratic but peak ventilation rate never coincides with peak wind speed. This house is reasonably 'tight' and during this period no space heating was operated, resulting in a smaller than typical temperature differential between the indoor air and that outside.

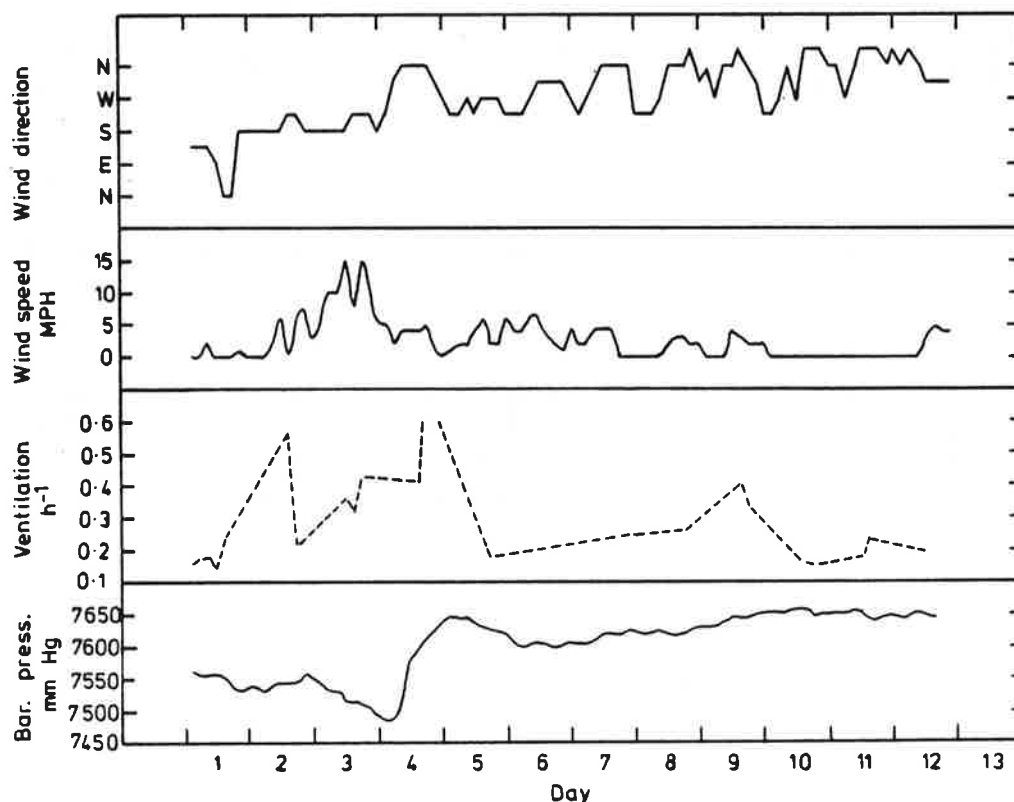


Figure 3 Wind speed, wind direction, barometric pressure and ventilation rate for house studied for 13 days

Figure 4 displays the results of the determination of the concentrations of the decay products of radon-222 in the lounge during this period, whereas Figure 5 shows the variation in the mWL value in the lounge. The decay products

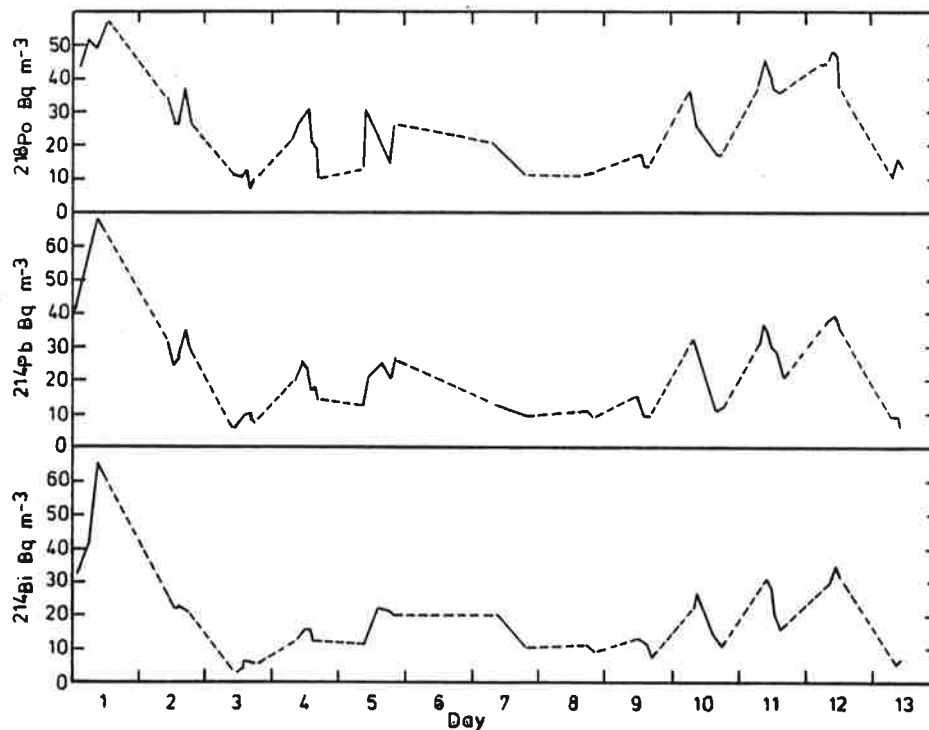


Figure 4  $^{222}\text{Rn}$  decay products in the lounge of house studied for 13 days

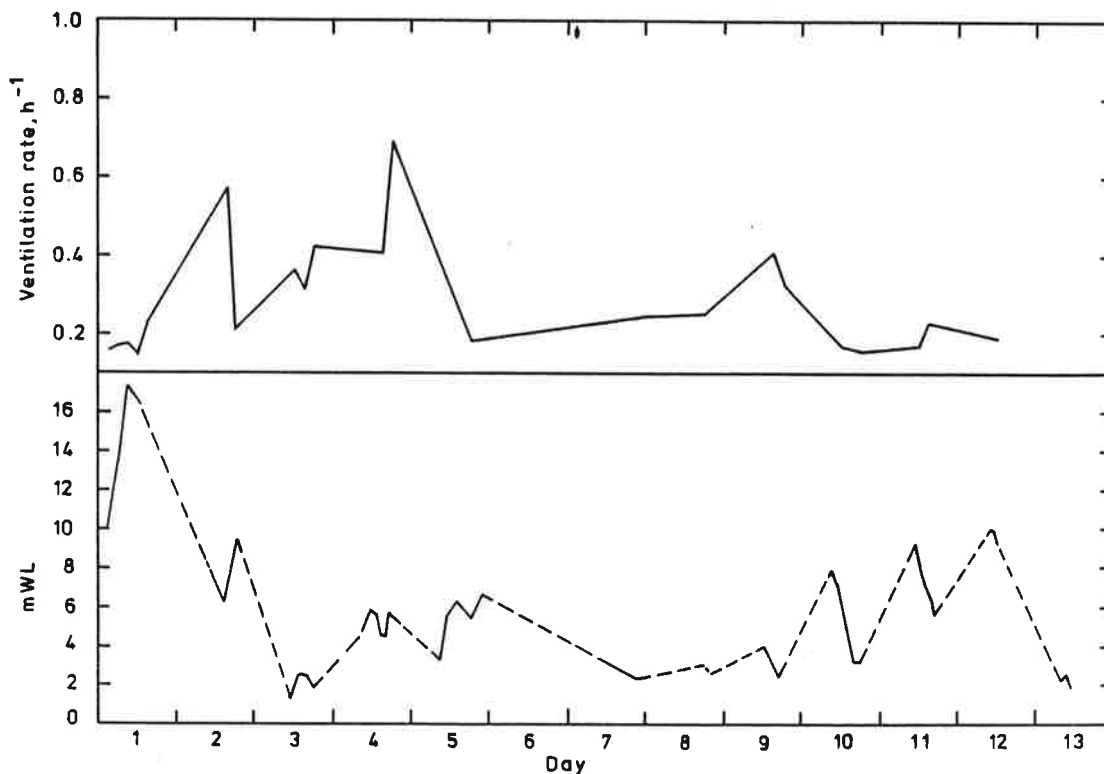


Figure 5 mWL due to  $^{222}\text{Rn}$  decay products in the lounge of house studied for 13 days

concentrations broadly follow an inverse trend to that of the ventilation rate, but the most obvious relationship is that between the mWL and the ventilation rate. This is aided in part because the mWL is determined with the greatest precision: typically 5% or better. Error bars have not been shown on the figures as in many cases they would overlap and be difficult to identify. As an indication of the order of the uncertainties on the measurements a few results are reproduced in Table 26.

Table 26  
Selected results of measurements of the decay products of  
radon-222 in the lounge

Day	Activity concentration (Bq m <sup>-3</sup> )			mWL
	Polonium-218	Lead-214	Bismuth-214	
1	43.5 ± 5.9	40.1 ± 2.1	33.5 ± 2.8	10.1 ± 0.2
3	10.7 ± 2.2	5.8 ± 0.74	2.94 ± 0.98	1.39 ± 0.06
5	13.7 ± 3.4	13.0 ± 1.2	11.9 ± 1.6	3.35 ± 0.10

Figures 6 and 7 show the temporal variation in the decay products concentration in the main bedroom. These are similar to those for the lounge

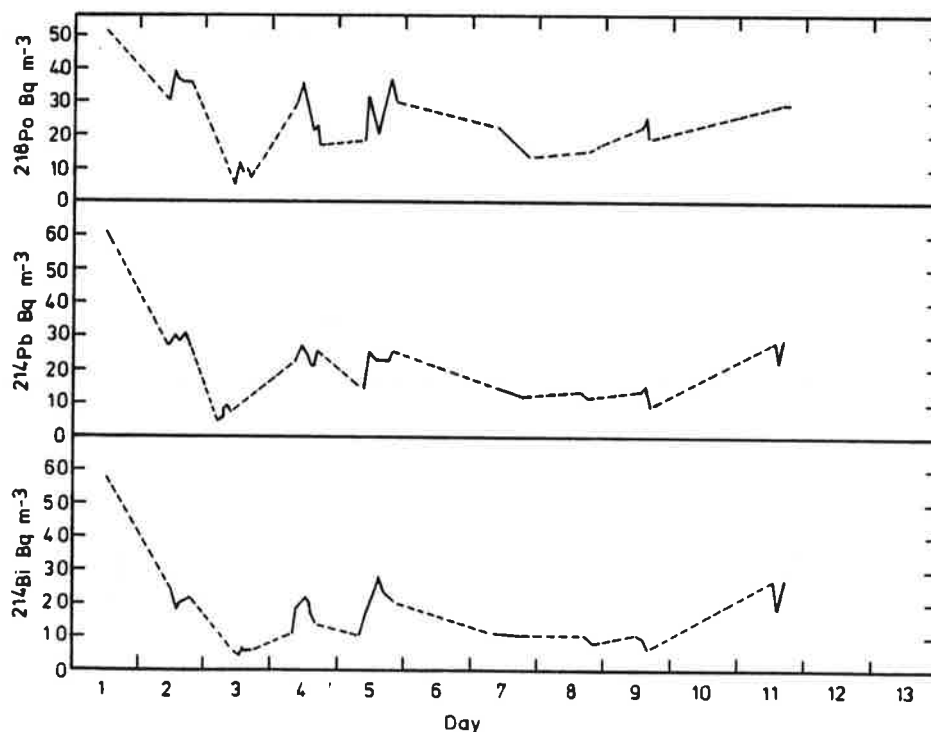


Figure 6 <sup>222</sup>Rn decay products concentrations in the main bedroom of the house studied for 13 days

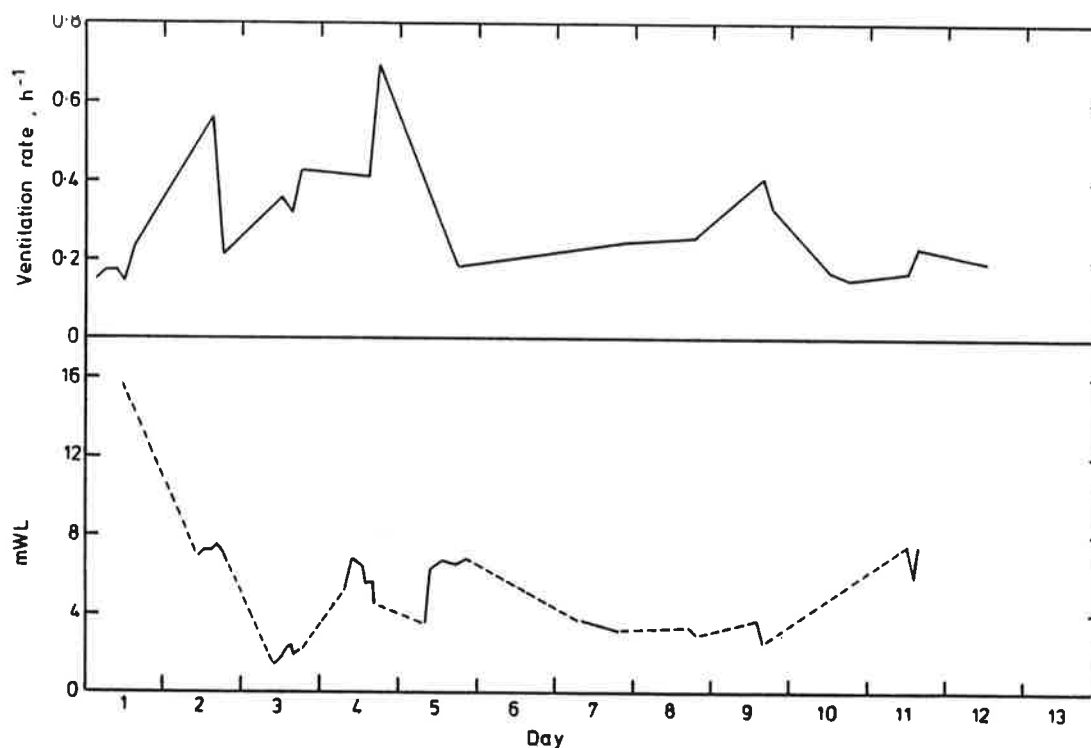


Figure 7 mWL values in the main bedroom and ventilation rate of the house studied for 13 days

shown in Figures 4 and 5. The results for the main bedroom tend to indicate slightly higher concentrations on the first floor than on the ground. However, the differences are not statistically significant when the uncertainties in the measurements are considered.

Throughout the measurement period the condensation nucleus (CN) concentration was monitored in the lounge and was found to be in the range  $8 \cdot 10^3$  to  $7 \cdot 10^4 \text{ ml}^{-1}$ . On day 10 the lounge door was partially closed and an electrostatic precipitator switched on at its maximum setting at 09.20. The precipitator was switched off at 14.26 and the lounge door opened wide at this time. Radon-222 decay products concentrations in the lounge were measured throughout the day. The effects of the electrostatic precipitator are shown in Figure 8. To avoid confusion, error bars are not shown on Figure 8 but the full results are given in Table 27.

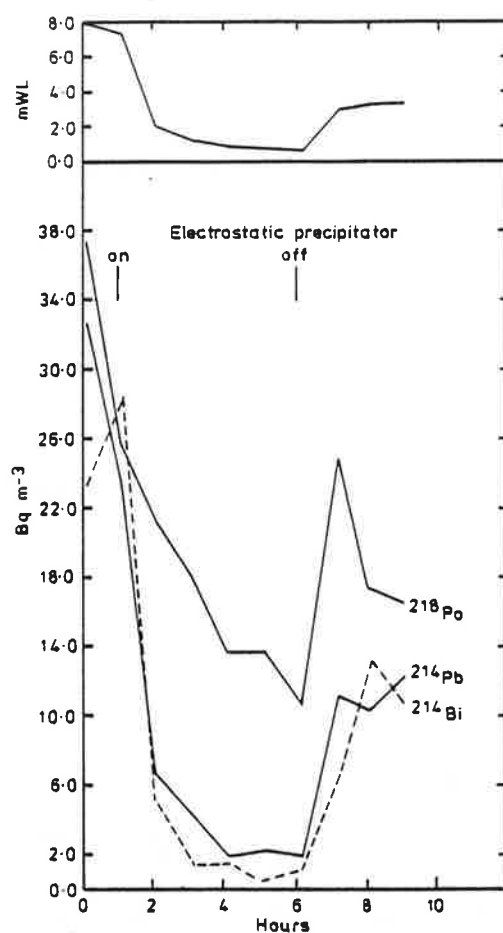


Figure 8 Effect of an electrostatic precipitator on the concentration of the decay products of  $^{222}\text{Rn}$  in room air

Table 27

Effect of electrostatic precipitator on the concentration of radon-222 decay products in air

Time	CN concentration	Activity concentration ( $\text{Bq m}^{-3}$ )			mWL
		Polonium-218	Lead-214	Bismuth-214	
08.15		$37.3 \pm 5.1$	$32.6 \pm 1.8$	$23.4 \pm 2.4$	$7.86 \pm 0.15$
09.15	$9.3 \cdot 10^3$	$25.8 \pm 5.0$	$27.3 \pm 1.75$	$27.9 \pm 2.4$	$7.27 \pm 0.14$
10.10	$2.0 \cdot 10^3$	$21.3 \pm 2.7$	$6.59 \pm 0.89$	$5.08 \pm 1.20$	$2.01 \pm 0.07$
11.20	$1.0 \cdot 10^2$	$18.1 \pm 2.13$	$4.20 \pm 0.68$	$1.44 \pm 0.90$	$1.22 \pm 0.06$
12.15	$5.19 \cdot 10^1$	$13.7 \pm 1.82$	$1.96 \pm 0.54$	$1.57 \pm 0.75$	$0.81 \pm 0.04$
13.15	$8.0 \cdot 10^1$	$13.7 \pm 1.68$	$2.18 \pm 0.51$	$0.417 \pm 0.689$	$0.722 \pm 0.04$
14.15	$9.75 \cdot 10^1$	$10.7 \pm 1.62$	$1.81 \pm 0.49$	$1.05 \pm 0.67$	$0.651 \pm 0.04$
15.15	$3.72 \cdot 10^3$	$24.9 \pm 3.2$	$11.1 \pm 1.08$	$6.96 \pm 1.44$	$2.92 \pm 0.089$
16.15	$5.32 \cdot 10^3$	$17.4 \pm 3.47$	$10.3 \pm 1.16$	$13.2 \pm 1.58$	$3.22 \pm 0.09$



The electrostatic precipitator can be seen to be an effective means of reducing the radon decay products concentration in a domestic setting, thus verifying the results found earlier in an environmental chamber<sup>(60)</sup>. However, to reduce the radon decay products exposure generally it would be necessary to install an electrostatic precipitator in at least two rooms on each floor. The electrostatic precipitator, by reducing the aerosol concentration, increases the proportion of radon-222 decay products in the air that are unattached to particles and this results in a higher dose to lung tissue for a given WLM exposure. A second adverse effect of electrostatic precipitators is the production of ozone. A detailed study of all the effects of electrostatic precipitators is warranted before their use to reduce the radon-222 decay product concentration is recommended.

Periodic measurements of the radon decay products concentration in outside air were carried out. In the majority of cases the concentrations were so low that the uncertainty on the determination of each nuclide concentration exceeded 50% of the calculated value (for polonium-218 this was often in excess of 100%). The mWL assessment, however, was subject to a worst-case uncertainty of 17%. The measurements were made at a height above the ground of approximately 1 m, and under most conditions the decay products at this height will be close to equilibrium. Table 28 gives the measured values of mWL in outside air and the corresponding equilibrium equivalent radon-222 concentration. The results for days 10, 11 and 12 show the typical feature of high values early in the morning declining as the day advances. The high mWL value during the afternoons of days one and two are unusual. That on day one probably results from the still weather conditions, but on day two the wind was moderate and there had been heavy rain overnight with intermittent light rain until 15.30. Heavy rain would be expected to 'washout' radon decay products and reduce the mWL. In all cases the high mWL value in outside air is reflected by a peak in the measured value indoors.

Measurements of the decay products of radon-220 as well as those of radon-222 were made from the same air sample on most days. These results are given in Table 29. There is a tendency for the ratio of the mWL of radon-220 to that of radon-222 to increase as the ventilation rate increases in line with the predictions by Stranden<sup>(59)</sup>, but no firm conclusions can be drawn as the range of ventilation rates is small, producing a cluster of values of the ratio for ventilation rates in the range 0.17–0.33 h<sup>-1</sup>.

The two higher ventilation rates indicate a higher ratio at 0.42 h<sup>-1</sup> than at 0.69 h<sup>-1</sup>. Further measurements are needed to demonstrate a true functional relationship.

Table 28

mWL in outside air and equilibrium equivalent radon-222 decay products  
concentration

Day	Time	mWL	Equilibrium equivalent radon-222 concentration <sup>1</sup> (Bq m <sup>-3</sup> )
1	15.30	1.42 ± 0.062	5.25
2	18.00	1.19 ± 0.017	4.40
3	14.00	0.086 ± 0.014	0.318
	15.00	0.077 ± 0.013	0.285
	17.00	0.097 ± 0.015	0.359
4	13.30	0.114 ± 0.016	0.422
	15.30	0.100 ± 0.015	0.370
5	18.15	0.102 ± 0.014	0.377
	19.15	0.125 ± 0.016	0.463
7	09.15	0.302 ± 0.029	1.12
	19.15	0.246 ± 0.026	0.910
8	18.00	0.224 ± 0.026	0.829
9	14.00	0.132 ± 0.019	0.488
	15.00	0.130 ± 0.019	0.481
10	07.45	1.13 ± 0.055	4.18
	11.00	0.431 ± 0.034	1.59
	13.45	0.249 ± 0.025	0.921
11	10.15	1.34 ± 0.061	4.96
	11.15	1.38 ± 0.062	5.11
	15.15	0.418 ± 0.034	1.55
12	07.30	3.07 ± 0.092	11.4
	08.30	2.31 ± 0.094	8.55
	09.30	1.82 ± 0.071	6.73
	11.30	0.637 ± 0.043	2.36
13	10.20	0.183 ± 0.026	0.677

<sup>1</sup>The equilibrium equivalent radon-222 concentration is that concentration of radon-222 in equilibrium with its short-lived decay products that would produce the observed WL.

Air samples were taken during this period with Lucas cells to determine the radon-222 gas concentration. Unfortunately the concentrations were too low for meaningful results to be obtained and it was not possible to assess the influence of plate-out as a mechanism for the removal of decay products from room air.

Table 29  
Radon-222 and radon-220 decay products concentrations

Day	Room	Activity concentration (Bq m <sup>-3</sup> )			mWL Radon-222	Activity concentration (Bq m <sup>-3</sup> )		mWL Radon-220	R	Ventilation rate (h <sup>-1</sup> )
		Polonium-218	Lead-214	Bismuth-214		Lead-212	Bismuth-212			
2	L	26.6±4.9	29.2±1.6	25.9±1.8	7.44±0.28	0.701±0.119	0.421±1.10	2.35±1.92	0.316±0.029	0.225
3	L	10.0±2.56	7.71±1.20	6.07±1.05	1.97±0.24	0.459±0.105	0.133±0.953	1.52±0.10	0.770±0.121	0.423
3	B	8.64±2.26	8.40±1.15	5.88±0.96	2.00±0.23	0.620±0.103	1.36±0.933	1.59±0.15	0.780±0.113	0.423
4	L	9.96±3.46	14.3±1.12	13.4±1.31	3.63±0.23	0.388±0.098	0.432±0.899	1.46±0.16	0.403±0.050	0.690
4	B	16.6±3.7	16.8±1.3	14.5±1.4	4.27±0.24	0.494±0.103	1.495±0.956	2.18±0.69	0.510±0.049	0.690
5	L	25.9±4.6	27.4±1.5	20.7±1.72	6.63±0.26	0.561±0.109	0.417±1.02	2.14±0.180	0.323±0.031	0.180
5	B	30.8±4.6	26.7±1.48	21.7±1.70	6.78±0.27	0.619±0.113	0.779±1.06	2.44±0.187	0.363±0.031	0.180
7	L	11.2±2.8	9.71±1.04	7.57±1.08	2.43±0.20	0.296±0.781	0.416±0.781	1.05±0.136	0.431±0.065	0.242
7	B	13.6±3.0	11.7±1.1	10.3±1.1	3.04±0.21	0.366±0.091	0.447±0.824	1.10±0.143	0.361±0.052	0.242
8	L	11.8±2.9	9.86±1.02	9.77±1.10	2.69±0.19	0.390±0.082	0.181±0.759	1.28±0.131	0.476±0.059	0.253
8	B	16.9±3.0	11.7±1.09	8.62±1.14	2.97±0.21	0.333±0.089	0.149±0.815	1.18±0.143	0.397±0.055	0.253
9	L	13.7±2.8	9.19±1.01	7.89±1.06	2.46±0.19	0.293±0.083	0.111±0.754	0.966±0.143	0.392±0.060	0.331
9	B	19.0±2.9	9.19±1.01	7.07±1.09	2.53±0.19	0.240±0.081	1.11±0.749	1.19±0.13	0.471±0.063	0.331
10	L	16.5±3.4	12.2±1.1	10.7±1.2	3.25±0.20	0.228±0.083	1.06±0.77	1.17±0.138	0.359±0.048	0.170
11	L	24.8±4.0	19.7±1.2	15.6±1.5	5.02±0.22	0.235±0.093	1.02±0.856	1.22±0.154	0.242±0.033	0.233

L = lounge; B = bedroom; R = ratio mWL radon-220/mWL radon-222.

6. THE THEORETICAL APPROACH TO THE TRANSPORT OF RADON THROUGH MATERIALS AND OF RADON DECAY PRODUCTS IN ROOM AIR

The migration of radon isotopes through a porous medium has been described as a three-step process<sup>(61)</sup>. The steps are:

- (i) the radioactive formation and recoil of the newly formed ion from its parent radium isotope;
- (ii) the diffusion of the neutral atom through the interior of a mineral grain;
- (iii) the diffusion and transport of the neutral atom through the pore space.

Since the diffusion coefficient of a gas in a mineral is of the order of  $10^{-27}$   $\text{cm}^2 \text{ s}^{-1}$  or less, only those atoms which terminate their recoil in a pore space or very close to the surface of a mineral grain are available for diffusion and transport through the pore spaces. Thus, only a small fraction of the atoms of radon formed within a material is ultimately released from the surface. This fraction is termed the emanating fraction or the emanating power and values for most building materials lie in the range 1-15%, as is shown in Tables 8 and 11 of Section 4. The exception in Table 8 is phosphogypsum, having mean values of emanating power exceeding 20%.

It is generally assumed that the migration of radon through walls and soils is dominated by the diffusion mechanism. Tanner<sup>(61)</sup> has suggested that the influence of transport is generally obscured by the use of an 'effective' diffusion coefficient and by uncertainties in the values of experimentally determined diffusion coefficients. Transport effects can be seen, however, in the variation of the radon exhalation rate with atmospheric pressure<sup>(37)(62)</sup>. It has been noted that the occurrence of a pressure change results in an inverse change in the radon exhalation from soil and from walls. Kraner<sup>(62)</sup> interpreted this effect as a short-range displacement of pore gas moving under the influence of a pressure differential between the atmosphere and the pore gas at depth.

Culot et al<sup>(39)</sup> have noted that the wide range of reported values for the diffusion coefficient of radon in particular materials may reflect ambiguity in the definition of the effective diffusion coefficient. In comparison to free diffusion, two factors reduce the flux of a substance across a reference volume filled by a porous medium:

- (a) the cross sectional area through which diffusion can take place is reduced by a fraction equal to the ratio of the open pore area to the total cross-section of the reference area;
- (b) the pathway imposed on the diffusion process by the circuitous nature of the pores is a lengthened one.

The flux of the gas may be expressed as

$$J = - K_e \text{ grad } C \quad \text{.....(5)}$$

where  $J$  is the gas flux per unit area;

$K_e$  the effective diffusion coefficient of the gas  
in the porous medium

and  $C$  the gas concentration per unit volume of interstitial space.

If the flux is expressed per unit area of open pores, only the lengthening of the pathway is taken into account and a larger value of the effective diffusion coefficient,  $K_e^*$ , will result. If the same flux is expressed per unit area of porous medium, a lower value,  $K_e$ , is obtained. Culot goes on to show that the diffusion equation resulting from the second definition,  $K_e$ , will necessarily contain an explicit reference to the porosity of the medium and that the two coefficients are related, thus

$$K_e = PK_e^* \quad \text{.....(6)}$$

where  $P$  is the porosity.

The migration of radon through soil towards the air-earth interface is generally represented by the one-dimensional diffusion equation<sup>(62)</sup>

$$\frac{dC(x)}{dt} = \frac{K_e}{P} \frac{d^2C(x)}{dx^2} - \lambda C(x) + f \quad \text{.....(7)}$$

where  $C$  is the activity concentration per unit volume pore space

$\lambda$  the radioactive decay constant of radon

$f$  the production of radon per unit volume of interstitial space

and  $x$  the distance from the air-earth interface.

The soil may be treated as a semi-infinite medium, so that the boundary conditions are  $C \rightarrow 0$  at  $x = 0$  and  $C_0 = C$  at  $x = \infty$ . The steady-state ( $dC/dt = 0$ ) solution of equation (7) under these boundary conditions is then

$$C = C_0 \{1 - \exp[-(\lambda P/K_e)^{1/2} x]\} \quad \text{.....(8)}$$

and the flux (from equation (5)) is given by

$$J = -K_e C_0 (\lambda P/K_e)^{1/2} \exp[-(\lambda P/K_e)^{1/2} x] \quad \text{.....(9)}$$

This solution has been applied to the radon flux from uranium mill tailings<sup>(63)</sup> and also to the radon flux from walls<sup>(64)</sup>.

However, a more realistic solution to equation (7) for the diffusion of radon through walls is that developed by Krisiuk et al<sup>(26)</sup> and by Jonassen and McLaughlin<sup>(27)</sup>. If the central point of the wall is taken as the origin then, by symmetry, the radon flux is equal to zero at  $x = 0$ , ie

$$-K_e \frac{dC}{dx} \Big|_{x=0} = 0 \quad \text{.....(10)}$$

For diffusion occurring from a wall  $2d$  thick into a room of unity depth at

the steady state ( $dC/dt = 0$ ), the flux into the room equals the radioactive decay

$$Ke \frac{dC}{dx} \Big|_{x=d} = C(d) \lambda \quad \text{.....(11)}$$

The steady-state solution of (7) under the boundary conditions (10) and (11) has the form

$$C(x) = \frac{f}{\lambda} \left[ 1 - \frac{\cosh (x/\ell_o)}{\cosh \beta + (\sinh \beta)/(\alpha\beta)} \right] \quad \text{.....(12)}$$

where  $\ell_o = (Ke/\lambda P)^{1/2}$  is the diffusion length

$\alpha = 1/dP$  the ratio of external to internal air volumes

$\beta = d/\ell_o$

and  $f = C_{Ra} (\lambda \rho \eta / P)$ ,

with  $C_{Ra}$  the activity concentration of radium in the material

$\rho$  the density

and  $\eta$  the fraction of radon-222 formed within the material that enters the pore spaces.

The flux of radon into the room is then given by

$$J = - Ke \frac{dC}{dx} \Big|_{x=d} = f P \ell_o \left[ \frac{\tanh \beta}{1 + (\tanh \beta)/(\alpha\beta)} \right] \quad \text{.....(13)}$$

For the case where the external volume is considerably greater than the volume of intra-wall air (ie,  $\alpha$  is large), the flux is given by

$$J = f P \ell_o \tanh \beta \quad \text{.....(14)}$$

This is a valid approximation for radon diffusion into a well-ventilated room since the effective external volume is substantially higher than the volume of the room. Thus, for most rooms, the exhalation rate is virtually independent of the ventilation rate.

If the fraction, of the radon entering the pore spaces that ultimately escapes from the surface of the material, is  $g$ , then  $(g\eta)$  is the emanation coefficient defined earlier.

The maximum value of  $J$  corresponds to the exhalation of all emanating atoms entering the pores

$$J_{\max} = C_{Ra} \eta \rho \lambda d \quad \text{.....(15)}$$

and thus

$$J = \frac{J_{\max}}{\beta} \tanh \beta \quad \text{.....(16)}$$

These solutions have been used to model exhalation rates and radon concentration in rooms, and to calculate the effective diffusion coefficient for various materials from measured exhalation rates<sup>(26,27,39,59,65)</sup>. In general, the results obtained by modelling are in good agreement with measured values.

The variation of the radon activity concentration in a room with time (assuming instantaneous mixing) is of the form<sup>(26)</sup>

$$\frac{dC}{dt} = J \frac{S}{V} + C_{in} j - C (\lambda + j) \quad \dots\dots(17)$$

where C is the activity concentration of radon (Bq m<sup>-3</sup>)  
 J the exhalation rate of radon from room surfaces (Bq m<sup>-2</sup> h<sup>-1</sup>)  
 S the area of exhaling surfaces (m<sup>2</sup>)  
 V the volume of room (m<sup>3</sup>)  
 C<sub>in</sub> the activity concentration of radon in the incoming air (Bq m<sup>-3</sup>)  
 λ the decay constant of radon (h<sup>-1</sup>)  
 and j the ventilation or air exchange rate (h<sup>-1</sup>).

Extra terms may be added to allow for the release of radon from other sources such as water or natural gas. Radon flux from the soil or fill around and under the building, as has already been remarked, is often the dominant source of radon within buildings. In regions with enhanced uranium mineralisation the contribution of radon from the subjacent soil often exceeds, by an order of magnitude, that from the fabric of the building.

The solution of (17) is of the form

$$C = \frac{J(S/V) + C_{in} j}{\lambda + j} \{1 - \exp[-(\lambda + j)t]\} + C_0 \exp[-(\lambda + j)t] \quad \dots\dots(18)$$

and at equilibrium (t → ∞)

$$C_{eq} = \frac{J(S/V) + C_{in} j}{\lambda + j} \quad \dots\dots(19)$$

For all occupied buildings j >> λ and equation (19) reduces to

$$C_{eq} = \frac{J(S/V)}{j} + C_{in} \quad \dots\dots(20)$$

Thus, the radon concentration indoors is inversely proportional to the ventilation rate. Apart from the previously mentioned effect of pressure changes on the radon exhalation rate, the variations in the radon concentration in a building are mainly dependent on the variations of the ventilation conditions. Ventilation rates vary because of meteorological conditions, such as wind and temperature, and by human activities such as opening or closing windows and doors. Stranden and Berteig<sup>(28)</sup> have shown a linear relationship between wind speed and ventilation rate, and between the ventilation rate and the temperature difference between indoor and outdoor air. In the measurements reported in Section 5.2.5, at one house there was only a marginal relationship between wind speed and ventilation rate, for 13 days, but this may have resulted from the low ventilation rates obtaining when all outside doors and windows were closed. In general, the radon concentration in a building is expected to fall with increasing wind speed and with increasing temperature gradient.

Radon itself contributes little to the radiation dose to lung tissue<sup>(12)</sup> and

it is the decay products of radon-222 that are of importance. The decay products of radon-222 are rarely in equilibrium with the parent gas, and the concept of equilibrium factor has therefore been introduced. The equilibrium factor, F, is defined as that concentration of radon-222 which, in equilibrium with its short-lived decay products, would produce the observed WL value, divided by the actual concentration of radon-222 in the air.

The original definition of the WL was based on  $3.7 \text{ kBq m}^{-3}$  ( $100 \text{ pCi l}^{-1}$ ) of radon-222 in equilibrium with its short-lived decay products and this produces  $1.278 \cdot 10^5 \text{ MeV l}^{-1}$  of potential alpha energy. This was then rounded to  $1.3 \cdot 10^5 \text{ MeV l}^{-1}$ , and this now defines the WL.  $1.3 \cdot 10^5 \text{ MeV l}^{-1}$  would be produced by  $3.763 \text{ kBq m}^{-3}$  ( $101.7 \text{ pCi l}^{-1}$ ) of radon-222 in equilibrium with its short-lived decay products.

Strictly, F, is given by

$$F = \frac{3763(WL)}{C_{Rn}} \quad \text{.....(21)}$$

where  $C_{Rn}$  is the radon-222 concentration ( $\text{Bq m}^{-3}$ )  
and WL the working level value.

However, by common usage, F is usually taken as

$$F = \frac{3700(WL)}{C_{Rn}} \quad \text{.....(22)}$$

or

$$F = \frac{100(WL)}{C_{Rn}^1} \quad \text{.....(23)}$$

where  $C_{Rn}^1$  is the radon-222 concentration in  $\text{pCi l}^{-1}$ .

For the decay products polonium-218, lead-214 and bismuth-214, the corresponding equation to (19) is

$$C_i = \frac{\lambda_{i-1} C_{i-1} + j C_{i,in}}{j + \lambda_i} \quad \text{.....(24)}$$

where  $i = 0$  is radon-222,  $i = 1$  is polonium-218,  $i = 2$  is lead-214,  $i = 3$  is bismuth-214

$C_i$  the activity concentration of nuclide i

$C_{i,in}$  the activity concentration of nuclide i in the incoming air

and  $\lambda_i$  the decay constant of nuclide i.

These values, when weighted by the contribution per unit activity concentration of each nuclide to the WL, can be summed to give the WL (see Appendix A). The weighting factors are:

$2.786 \cdot 10^{-5} \text{ WL per Bq m}^{-3}$  polonium-218

$1.370 \cdot 10^{-4} \text{ WL per Bq m}^{-3}$  lead-214

$1.008 \cdot 10^{-4} \text{ WL per Bq m}^{-3}$  bismuth-214.

The equilibrium factor is then readily calculated for a particular ventilation



rate, eg,  $F = 0.54$  for  $j = 1 \text{ h}^{-1}$  if the concentrations of the decay products in the incoming air are assumed to be zero.

However, this is only a theoretical value of  $F$  based on the assumption that the decay products are lost from room air solely by radioactive decay and ventilation. In practice some of the airborne decay products will be lost from room air by attachment to surfaces within the room. This removal mechanism is generally termed plate-out. Thus, the radon decay-product concentration in room air will generally be less than that calculated above. The deposition rate of freshly formed atoms that have yet to attach themselves to aerosol particles is much greater than that for atoms attached to aerosol particles. In practice, the decay product most likely to be in the unattached state is polonium-218. Jacobi<sup>(66)</sup> has developed a theoretical model which accounts for the effects of aerosol concentration and the deposition rates of unattached and attached atoms on the radon decay products concentration. For a room with rough surfaces and a surface to volume ratio of  $2 \text{ m}^{-1}$  the plate-out rate constants for unattached and attached decay products are 70 and  $0.7 \text{ h}^{-1}$ , respectively. If it is assumed that all the lead-214 and bismuth-214 atoms are attached to aerosol particles, but a fraction,  $a$ , of polonium-218 is unattached, then the equilibrium factor using the plate-out rate constants above is given by (assuming that the concentrations of the decay products in the incoming air are zero)<sup>(67)</sup>

$$F = \frac{0.103\lambda_A}{(\lambda_A + j + 0.7 + 69.3a)} + \frac{0.507\lambda_A\lambda_B}{(\lambda_A + j + 0.7 + 69.3a)(\lambda_B + 0.7 + j)} + \frac{0.373\lambda_A\lambda_B\lambda_C}{(\lambda_A + j + 0.7 + 69.3a)(\lambda_B + j + 0.7)(\lambda_C + j + 0.7)} \dots(25)$$

where  $\lambda_A$  is the decay constant of polonium-218

$\lambda_B$  the decay constant of lead-214

$\lambda_C$  the decay constant of bismuth-214

and  $j$  the ventilation rate.

Some measurements of the equilibrium factor have been made during surveys of radon decay product concentrations in dwellings, but in many instances the radon-222 gas concentration was too low to obtain adequate precision on the radon-222 concentration using the Lucas flask method (see Appendix B). Thus, when taking the ratio of the WL determination to the radon-222 concentration, the uncertainty in the calculated value of  $F$  is unacceptable. Most results would indicate that the unattached fraction of polonium-218 is between 0 and 10%. At a ventilation rate of one air change an hour and with 10% unattached polonium-218,  $F$  becomes 0.28 rather than the 0.54 when plate-out is ignored. Experiments carried out in an environmental chamber have, however, indicated that Jacobi's model overestimates the removal of radon decay products from room air<sup>(68)</sup>. Further studies of the plate-out phenomena will be undertaken when more sensitive

equipment for the measurement of the concentration in air of radon-222 is available.

The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR)<sup>(69)</sup> have suggested that the theoretical value of the equilibrium factor (eg,  $F = 0.54$  at  $j = 1$ ) should be multiplied by 0.5 to account for plate-out. Measurements reported here would definitely indicate that the modifying factor of 0.5 is too small.

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## Appendix A

### The natural isotopes of radon and some common units of measurements

Before discussing the natural isotopes of radon and their decay products it may be useful to define some terms commonly used in radiological protection. In line with other scientific disciplines, radiological protection has adopted SI units of measurement, but the use of the new units is, as yet, far from universal. Consequently, much of the modern literature is written using the old, or traditional units. In the definitions below the old units are referred to as traditional units.

The activity,  $A$ , of an amount of radioactive nuclide in a particular energy state at a given time is the quotient of  $dN$  by  $dt$ , where  $dN$  is the expectation value of the number of spontaneous nuclear transitions from that energy state in the time interval  $dt$

$$A = dN/dt \quad \text{..... (A1)}$$

The special name for the SI unit is the becquerel (Bq) and the basic unit is  $s^{-1}$

$$1 \text{ Bq} = 1 \text{ s}^{-1}.$$

The curie (Ci) is the traditional unit for activity

$$1 \text{ Ci} = 3.7 \cdot 10^{10} \text{ s}^{-1} \text{ (exactly); } 1 \text{ Bq} = 2.7 \times 10^{-11} \text{ Ci}.$$

In environmental radioactivity the pCi ( $10^{-12}$  Ci) is commonly used

$$1 \text{ Bq} = 27 \text{ pCi}; 1 \text{ pCi} = 0.037 \text{ Bq}.$$

In terms of activity concentration

$$1 \text{ pCi l}^{-1} = 37 \text{ Bq m}^{-3}.$$

Absorbed dose,  $D$ , is the quotient of  $d\bar{e}$  by  $dm$ , where  $d\bar{e}$  is the mean energy imparted by ionising radiation to matter of mass  $dm$

$$D = d\bar{e}/dm \quad \text{..... (A2)}$$

The basic unit is  $\text{J kg}^{-1}$  and the special name for the SI unit of absorbed dose is the gray (Gy)

$$1 \text{ Gy} = 1 \text{ J kg}^{-1}.$$

The traditional name for the unit of absorbed dose is the rad

$$1 \text{ rad} = 10^{-2} \text{ J kg}^{-1}; 1 \text{ gray} = 100 \text{ rad}.$$

Dose equivalent is the quantity obtained by multiplying the absorbed dose by a factor (dimensionless) to allow for the different effectiveness of the various ionising radiations in causing harm to tissue. The factor for gamma rays, X-rays and beta particles is 1, for neutrons, 10, and for alpha particles, 20.

The special name for the SI unit of dose equivalent is the sievert (Sv)

$$1 \text{ Sv} = 1 \text{ J kg}^{-1}.$$

The traditional name for the unit of dose equivalent is the rem

$$1 \text{ Sv} = 100 \text{ rem}.$$

Effective dose equivalent is a concept introduced by the International Commission on Radiological Protection in 1977<sup>(1)</sup>. In the early days of radiological protection the main emphasis was on the detriment accruing to an individual who was uniformly irradiated from head to foot, but in many instances only a part of the body is irradiated. For the range of dose equivalents normally encountered in radiological protection the irradiation of a limited part of the human body is less detrimental than uniform whole body irradiation. The effective dose equivalent, therefore, applies to one or more organs and is the whole body dose equivalent which would produce the same total detriment to the individual as is produced by the actual dose equivalent received by the specific organ or organs. For a number of organs, ICRP have produced weighting factors by which the dose equivalent received by that organ is to be multiplied to give the effective dose equivalent. For lung the weighting factor is 0.12, and hence if the lung alone receives a dose equivalent of 1 Sv the effective dose equivalent is 0.12 Sv, implying that the same detriment results from 0.12 Sv received uniformly by the whole body as does a dose equivalent of 1 Sv to the lungs alone.

There exists in nature three series of radioactive nuclides that are termed primordial. For a radionuclide to occur naturally in the earth's crust it must either have a half-life comparable with, or longer than, the age of the earth, or it must itself be formed as the result of some other nuclide having the requisite half-life. Other naturally occurring radionuclides are formed by the interaction of cosmic rays with elements in the atmosphere, but these are of no concern here.

The three primordial series are thorium, uranium and actinium, and in the decay scheme of each series there occurs an isotope of radon. The members of the respective decay schemes, down to the corresponding radon isotope, are shown in Tables A1, A2 and A3, respectively.

Table A1  
The thorium series

Nuclide		Mode of decay	Half-life (y = year, d = day, h = hour, s = second)
Element	Isotope		
Thorium	Thorium-232	$\alpha$	$1.39 \cdot 10^{10}$ y
Radium	Radium-228	$\beta$	6.7 y
Actinium	Actinium-228	$\beta$	6.13 h
Thorium	Thorium-228	$\alpha$	1.91 y
Radium	Radium-224	$\alpha$	3.64 d
Radon	Radon-220	$\alpha$	54.5 s



Table A2  
The uranium series

Nuclide		Mode of decay	Half-life (y = year, d = day, min = minute)
Element	Isotope		
Uranium	Uranium-238	$\alpha$	$4.50 \cdot 10^9$ y
Thorium	Thorium-234	$\beta$	24.1 d
Protactinium	Protactinium-234	$\beta$	1.18 min
Uranium	Uranium-234	$\alpha$	$2.5 \cdot 10^5$ y
Thorium	Thorium-230	$\alpha$	$8.0 \cdot 10^4$ y
Radium	Radium-226	$\alpha$	$1.62 \cdot 10^3$ y
Radon	Radon-222	$\alpha$	3.82 d

Table A3  
The actinium series

Nuclide		Mode of decay	Half-life (y = year, d = day, h = hour, min = minute, s = second)
Element	Isotope		
Uranium	Uranium-235	$\alpha$	$7.1 \cdot 10^8$ y
Thorium	Thorium-231	$\beta$	25.6 h
Protactinium	Protactinium-231	$\alpha$	$3.43 \cdot 10^4$ y
Actinium	Actinium-227	$\beta$	22.0 y
Thorium	Thorium-227	$\alpha$	18.17 d
Francium	Francium-223	$\alpha, \beta$	22.0 min
Radium	Radium-223	$\alpha$	11.68 d
Astatine	Astatine-219	$\alpha$	0.9 min
Radon	Radon-219	$\alpha$	3.92 s

It can be seen from Tables A2 and A3 that the half-life of uranium-235 is much shorter than that of uranium-238, and as a consequence, natural uranium at the present time contains only 0.71% by weight of uranium-235, the remainder being uranium-238 (with a minute trace of uranium-234). The specific activity of a radionuclide (the activity in Bq per unit mass of material) is inversely proportional to the half-life. In terms of activity, uranium-235 represents about 4.3% of the activity of natural uranium. For this reason the other members of the actinium series, including radon-219 (also known as actinon) and its descendents, do not pose a radiological hazard in the general environment because of the low production rate of radon-219 and its short half-life. In

uranium processing plants, exposure to radon-219 may need to be considered along with the other isotopes of radon. This is then an occupational exposure and is of no concern as an environmental exposure.

In various rocks the activity concentrations of uranium-238 and thorium-232 are often similar, and so are the solid members of each series down to and including the radium isotopes. The decay product of radium is the noble gas radon which, being gaseous, can migrate through pore spaces in soil and rocks and reach the atmosphere. However, any radon-220 formed at depth will decay before it reaches the surface because of its short half-life of 54.5 s. Radon-222, with a half-life of 3.82 d, can migrate a considerable distance before a significant proportion has decayed. It is therefore common in most environments to find a higher activity concentration of radon-222 than of radon-220, and for this reason most studies of human exposure to radon and its decay products have centred on the isotope radon-222. However, it has recently been shown that population exposures to the decay products of radon-220 may not be trivial, and the decay products of both radon-222 and radon-220 are treated here. Both isotopes of radon decay according to similar schemes, which are shown in Figure A1.

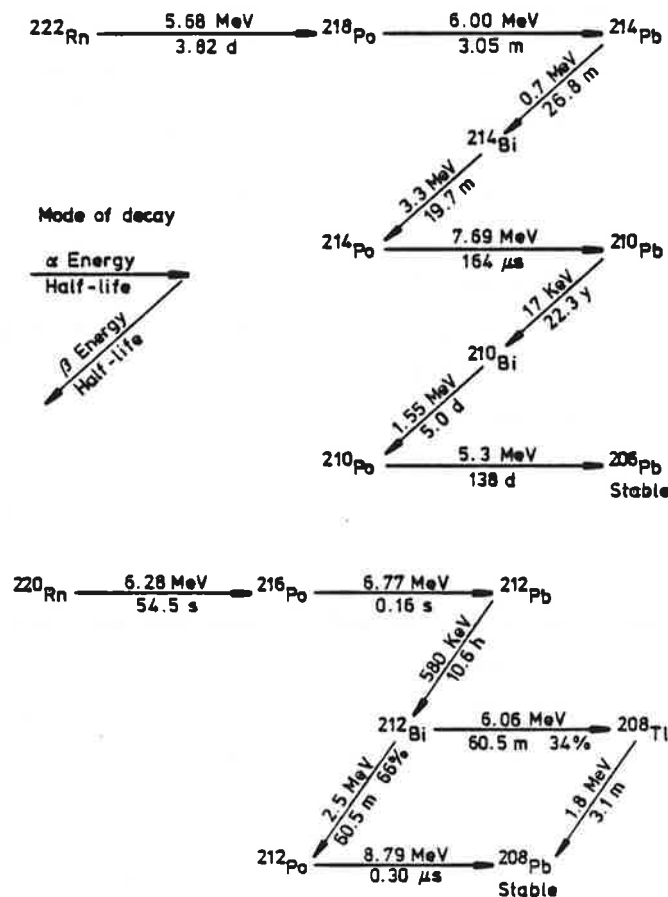


Figure A1 The decay schemes of  $^{220}\text{Rn}$  and  $^{222}\text{Rn}$

The decay schemes shown in Figure A1 are simplified by omitting those modes of decay which only occur as a fraction of a percent of the decay series.

As the deleterious effects of the inhalation of radon-222 and its decay products have been subjected to greater study than the corresponding hazards from radon-220, radon-222 and its decay products will be considered first. It should be noted that the decay products of radon-222 (polonium-218, lead-214, bismuth-214 and polonium-214) have been traditionally known as RaA, RaB, RaC and RaC<sup>1</sup>, respectively, and this nomenclature is still common in the literature.

As was noted in Section 2 of the main report a high excess incidence of lung cancer was observed among miners in Europe and North America who worked in mines where the ores had a high uranium content. Although this was first thought to be attributable to the high radon-222 gas concentrations in these mines it was conclusively demonstrated in 1951 that the radon-222 gas was relatively innocuous compared with its short-lived decay products<sup>(2)</sup>. From Figure A1 it can be seen that the half lives of these nuclides vary from 164  $\mu$ s for polonium-214 to a maximum of 26.8 minutes for lead-214. All these decay products of radon-222 are heavy metals and the freshly formed atoms become attached to aerosol particles that are inhaled. A fraction of the inhaled decay products will attach to surfaces lining the airways of the lung by diffusion, impaction and sedimentation. The two polonium isotopes decay by emitting alpha particles which result in high doses to small volumes of tissue. It is this irradiation of lung tissue by alpha particles that is thought to result in the increased incidence of lung cancer in miners exposed to radon-222 decay products.

The alpha-emitting decay products of radon-222 are polonium-218, polonium-214 and polonium-210. Polonium-210 is preceded by lead-210 which has a 22.3 y half-life and will be cleared from lung before a significant fraction decays. Thus, from a hazard point of view, the decay products of radon-222 are considered to end at polonium-214. In practice, the very short half-life of polonium-214 results in bismuth-214 appearing to behave as though it first emits a beta particle and shortly afterwards an alpha particle. This means that, in practice, bismuth-214 can be regarded as a combined beta and alpha-particle emitter. The short half-life of polonium-214 also ensures there being equal activity concentrations in air of bismuth-214 and polonium-214. Thus in air sampling, to determine the airborne concentration of the short-lived decay products, only concentrations of polonium-218, lead-214 and bismuth-214 need be determined.

At the time that it was first determined that the hazard in radon laden air was not radon-222, but the short-lived decay products of radon-222 in the air, no simple rugged equipment existed for determining the relative concentrations in air of polonium-218, lead-214 and bismuth-214. However, as it was thought that the alpha-particle irradiation of lung tissue would lead to lung cancer, an

indication of the hazard that a particular atmosphere posed might be given by the potential alpha-energy concentration in air. This can be determined with adequate precision by passing a sample of the air of interest through a filter paper, and at a later time determining the rate at which alpha particles are released by the dust collected on the filter paper.

At the time of these deliberations it was considered that 100 pCi l<sup>-1</sup> of radon-222 in equilibrium with its short-lived decay products (that is 100 pCi l<sup>-1</sup> each of polonium-218, lead-214, bismuth-214 and polonium-214) should correspond to the maximum permitted potential alpha-energy concentration in the air of uranium mines. This was the origin of the working level (WL) unit, the derivation of which is given in Table A4<sup>(3)</sup>.

Table A4  
The definition of the working level (WL)

Nuclide	Alpha-particle energy (MeV)	Half-life	Number of atoms per 100 pCi	Ultimate alpha particle per atom (MeV)	Total ultimate alpha-particle energy (MeV/100 pCi)
Radon-222	5.49	3.82 d	1.77 10 <sup>6</sup>	Excluded	None
Polonium-218	6.00	3.05 min	977	6.00+7.69	0.134 10 <sup>5</sup>
Lead-214	0	26.8 min	8580	7.69	0.660 10 <sup>5</sup>
Bismuth-214	0	19.7 min	6310	7.69	0.485 10 <sup>5</sup>
Polonium-214	7.69	164 $\mu$ s	0.0008	7.69	0.000 10 <sup>5</sup>
TOTAL					1.279 10 <sup>5</sup>
Rounded up to					1.3 10 <sup>5</sup>

Thus, 100 pCi l<sup>-1</sup> each of polonium-218, lead-214 and bismuth-214 will ultimately be responsible for the release of 1.279 10<sup>5</sup> MeV of alpha-particle energy, but to avoid unwarranted precision this value was rounded to 1.3 10<sup>5</sup> MeV. From Table A4 it will be seen that a polonium-218 atom will release 6.00 MeV of alpha-particle energy, when it decays to form a lead-214 atom. This lead-214 atom does not release any alpha-particle energy when it decays to form a bismuth-214 atom. The bismuth-214 atom in decaying to form polonium-214 again releases no alpha-particle energy but when the polonium-214 atom decays it releases 7.69 MeV of alpha-particle energy. The original polonium-218 atom is then responsible for the ultimate release of 6.00 + 7.69 = 13.69 MeV of alpha-particle energy and this is the contribution each polonium-218 atom in air makes to the WL. Similarly, each lead-214 and bismuth-214 atom in the air, although not themselves decaying by alpha-particle emission, will ultimately release 7.69 MeV of alpha-particle

energy as their subsequent decay products pass through the polonium-214 stage. Each atom of lead-214 and bismuth-214 in air then contributes 7.69 MeV to the WL.

The original basis of the WL was 100 pCi l<sup>-1</sup> and in SI units this becomes 3.7 kBq m<sup>-3</sup>, and the WL in SI units is defined as any combination of the short-lived decay products of radon-222 (polonium-218, lead-214, bismuth-214 and polonium-214) in 1 m<sup>3</sup> of air that will result in the ultimate release of 1.3 10<sup>8</sup> MeV of alpha-particle energy.

Note that, having derived the WL from set equilibrium conditions, the final definition is independent of the equilibrium conditions; the only constraint being on the ultimate alpha-particle energy concentration.

Table A5 presents a number of decay product concentrations which all produce 1 WL.

Table A5  
A selection of radon-222 decay products equilibrium  
conditions that yield 1 WL

Nuclide	Activity concentration (kBq m <sup>-3</sup> )					
Polonium-218	3.7	4.81	6.66	9.25	18.5	35.9
Lead-214	3.7	4.57	4.48	4.66	2.47	0
Bismuth-214	3.7	2.22	1.85	0.925	1.48	0

The working level is of course nothing more than the potential alpha-particle energy concentration in the air. A person breathing air of a given WL will receive a dose to lung tissue which will (for a steady breathing rate) increase linearly with the length of time spent in that atmosphere. Thus a unit of cumulative exposure was required, and this was derived in terms of the working month which was taken as 170 h. An exposure to 1 WL for 170 h produces a cumulative exposure of one working level month (WLM). Likewise, an exposure to 2 WL for 85 h, or to 0.5 WL for 340 h, also results in a cumulative exposure of 1 WLM.

In 1967 the maximum permissible annual exposure to radon-222 decay products in US mines was set at 12 WLM, but in 1969 this was reduced to 4 WLM<sup>(4)</sup>. In 1978 ICRP recommended a limit of 5 WLM<sup>(5)</sup>. The ICRP recommendation, however, takes into account the additivity of all effective dose equivalents. They have concluded that for workers, 1 WLM corresponds to an effective dose equivalent of 10 mSv. Since the total of all effective dose equivalents must not exceed 50 mSv in a year and the gamma dose received in many mines approaches 10 mSv in a year,

this would, in most instances, limit the permitted exposure to radon-222 decay products to 40 mSv or 4 WLM.

For exposures to radon-222 decay products in the general environment it should be apparent that the dose to lung tissue from a given exposure in WLM will be different from that applying in the mining situation, principally because breathing rates will be quite different. The relationship between the deposition of the decay products of radon-222 in lung and breathing rate are not simple. Generally, the lower the breathing rate the lower the dose rate to lung tissue when inhaling a given radon-222 laden atmosphere. For this and other reasons the conversion factor from WLM to effective dose equivalent for environmental exposures in favour in the UK at this time is 1 WLM = 5 mSv effective dose equivalent<sup>(6)</sup>.

The WL was specifically introduced to aid in assessing the hazard posed by inhaling an atmosphere containing the decay products of radon-222. However in some industries, including mining, materials rich in thorium-232 are handled; one such industry is the manufacture of gas mantles. In these circumstances workers can be exposed to high concentrations of radon-220 and its decay products. The WLM concept was in consequence extended for use in measuring exposure to the decay products of radon-220, but this move did not go unchallenged<sup>(7)</sup>. There are similarities in the decay schemes of radon-222 and radon-220 in that both chains contain polonium isotopes that emit alpha particles, but there are significant differences.

With the half-life of radon-222 being much greater than that of any of its first four decay products, these decay products are rarely very far from equilibrium with the parent gas. Under these circumstances nearly all the dose to lung tissue arises from the inhalation of the decay products of radon-222. Radon-220 has a half-life of 54.5 s and that of its first decay product polonium-216 a half-life of only 0.16 s. At any distance from a source of radon-220 the radon-220 and polonium-216 will have decayed, and the hazard arises from the inhalation of lead-212 and bismuth-212. Close to thorium ore bodies the concentrations of radon-220 and polonium-216 may be much higher (a factor of 100) than that of lead-212. In this case the most significant source of dose to lung tissue may be the radon-220 and polonium-216 rather than the later members of the decay scheme, lead-212 and bismuth-212. It was stated when discussing the origin of the WL that essentially, as the decay products of radon-222 had half lives less than 26.8 minutes, all these nuclides deposited in lung would decay there. In the case of lead-212, with a 10.64 h half-life, a substantial fraction of that deposited will be removed from lung by biological mechanisms. This results in the dose to lung tissue from an exposure of 1 WLM to the decay products of radon-220 being less than the corresponding dose from exposure of 1 WLM to the decay products of radon-222. However, lead-212 removed biologically from lung is moved to other body sites, and hence 1 WLM exposure to the decay products of radon-220

implies radiation doses, that are not negligible, to organs other than lung. Taking all these factors into account, ICRP<sup>(5)</sup> have stated that for workers the conversion factor from WLM, from radon-220 decay products, to effective dose equivalent should be one third that applicable to a WLM exposure to the decay products of radon-222. No other study has considered the conversion factor applicable to radon-220 decay product exposures in the general environment. However, there seems no sound reason to suppose that the factor of three suggested by ICRP for workers should be any different for exposures in the general environment. The appropriate relationship between a WLM exposure to the decay products of radon-220 (lead-212 and bismuth-212) and effective dose equivalent is then 1 WLM = 2 mSv, bearing in mind that ICRP does not consider that the basis of these conversion factors warrants more than one significant figure.

The activity concentrations of lead-212 and bismuth-212 to produce 1 WL are considerably smaller than for the decay products of radon-222. The derivation of the corresponding activity concentrations of lead-212 and bismuth-212 is approached in a similar manner to that outlined in Table A4, except that the starting point for the WL of radon-222 decay products was a given concentration of the decay products followed by a calculation of the potential alpha-particle energy. For the decay products of radon-220 the definition of the WL is taken as being the activity concentration necessary to produce a potential alpha-particle energy concentration of  $1.3 \times 10^8$  MeV in  $1 \text{ m}^3$  of air. Matters are slightly more complicated by the branching decay scheme for the routes taken from bismuth-212 to lead-208. In the total transition from bismuth-212 to polonium-208, 34% of the alpha particles emitted have an energy of 6.06 MeV and 66% an energy of 8.78 MeV. Thus, on average, the decay of a bismuth-212 atom will release 7.86 MeV of alpha-particle energy. Likewise, for each lead-212 atom that decays it too will release 7.86 MeV of alpha-particle energy when it passes through the bismuth-212 decay stage.

One Bq of lead-212 contains 55261 atoms and 1 Bq of bismuth-212 contains 5246 atoms. Hence, 1 Bq each of lead-212 and bismuth-212 contains 60507 atoms, each of which will ultimately release 7.86 MeV of alpha-particle energy, or a total of  $4.76 \times 10^5$  MeV of potential alpha-particle energy.

Thus the number of Bq each of lead-212 and bismuth-212 in  $1 \text{ m}^3$  of air to produce 1 WL is  $273 \text{ Bq m}^{-3}$ , or  $7.39 \text{ pCi l}^{-1}$ .

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Appendix B  
Measurement Techniques

1. DETERMINATION OF THE RADIONUCLIDE CONTENT OF MATERIALS

Naturally occurring radionuclides which are present in building materials and act as sources of exposure of the general public are thorium-232, radium-226 and their decay products, and potassium-40. Of all natural potassium, 0.012% is represented by the radioactive isotope potassium-40 which decays by emitting gamma rays. In principle, the potassium-40 content of a building material can be ascertained if the total potassium content is determined. It is more usual to determine the potassium-40 content by gamma-ray spectrometry at the same time that the thorium-232 and radium-226 content is determined. The method employed to determine the radium-226 and thorium-232 content is to grind the material to a size that ensures homogeneity in a specimen of approximately 900 cc. The specimen is then sealed in an air-tight container. When few specimens are to be handled, specially designed brass containers of regular geometry are used, but these containers are few in number and, as each specimen has to be stored in such a container for approximately 5 weeks before the measurement is carried out, a standard Kilner jar was used.

The reason for sealing the specimen is that the gamma rays detected are those from the decay products of radium-226 and thorium-232, including the nuclides beyond the respective radon isotopes. In the free state in which some of the radon isotopes are permitted to escape from the specimen, gamma-ray spectrometry would indicate a lower content of thorium-232 and radium-226 than the true value because only a fraction of the gamma-emitting nuclides are retained in the specimen. If a specimen is hermetically sealed in a container and left for about 5 weeks or longer no radon will escape, and radon and its short-lived decay products will be in equilibrium with the parent radium.

Standard radioactive sources are made up, containing precisely known activities of radium-226 and thorium-232, and the standard source containers are of similar geometry to that of the specimen container. A gamma-ray spectrum from each standard source is recorded and stored by computer. The specimen container containing the crushed material is then placed in the gamma-ray spectrometer and the spectrum that is obtained is compared with that obtained from the standard sources. Using a regression technique to compare the spectra, the activity of radium-226 and thorium-232 is determined. The potassium-40 content of a material can also be measured by this form of gamma-ray spectrometry. Unfortunately the Kilner jars used as storage containers had variable potassium contents in the glass, and hence the potassium-40 assessments could not be interpreted.

The limits of detection of the spectrometry method are approximately 1 Bq for radium-226 and thorium-232, and 5 Bq for potassium-40 if the correct container is used.

A more sensitive technique for radium-226 involves the total dissolution of part of the specimen. Subsequent chemical extraction of the radium-226 and alpha-particle counting of radon-222 in equilibrium with radium-226 provides a measurement technique sensitive to levels of  $4.10^{-4}$  Bq. Thorium-232 can be determined by ion exchange of the dissolved specimen and subsequent electroplating and alpha-particle counting. This method is sensitive to  $4.10^{-5}$  Bq. These latter methods have been used for a small number of specimens and agreement with results from spectrometry are good. However, the chemical methods are more time consuming than gamma-ray spectrometry and require the attention of skilled personnel.

## 2. THE DETERMINATION OF THE RATE OF EMANATION OF RADON-222 FROM MATERIALS

For many years the emanation rate of radon-222 from a material was determined by sealing the specimen into an airtight container of known volume. The specimen was left in the container for about 5 weeks. After this period had elapsed the rate of formation of radon-222 in the air space of the container would be equal to the rate of radioactive decay of the radon-222 in the container. The system is said to be in equilibrium and the concentration of radon in the container air is constant. By assessing the radon concentration in a small volume of the container air the rate of production of radon-222 by the specimen can be calculated.

However, it was pointed out<sup>(1)</sup> that this method will produce a substantial underestimate of the rate of emanation of radon-222 from the specimen surfaces when the specimen is emanating to air of zero or low radon-222 concentration. This effect was termed the 'back diffusion' effect and is a consequence of Fick's Law. For a material such as brick the release of radon-222 from the surface is the result, in part at least, of diffusion of radon-222 from the high concentration in the pore spaces near the centre of the brick to the low concentration in the air surrounding the brick. The rate of diffusion is proportional to the concentration gradient of radon-222. When the specimen is sealed in a container the concentration of radon-222 in the air surrounding the brick will increase and hence the rate of diffusion will decrease.

It is not essential that the measurement of radon-222 emanation from specimens should be attempted while the specimen is surrounded by radon-free air as this is a situation seldom met in practice. An adequate measure of the radon-222 emanation rate can be determined from a series of measurements of the concentration of radon-222 in the air surrounding the specimen before the radon-222 concentration in that air becomes high enough to seriously retard the emanation.

Figure B1 displays the system used in this report to determine the exhalation rate of radon-222 from building materials. The specimen is supported in a rectangular air-tight container. The container is fitted with a small carbon-vane air pump, the power leads of which are brought out through the sides of the container and sealing compound applied to make sure that there is no air leak

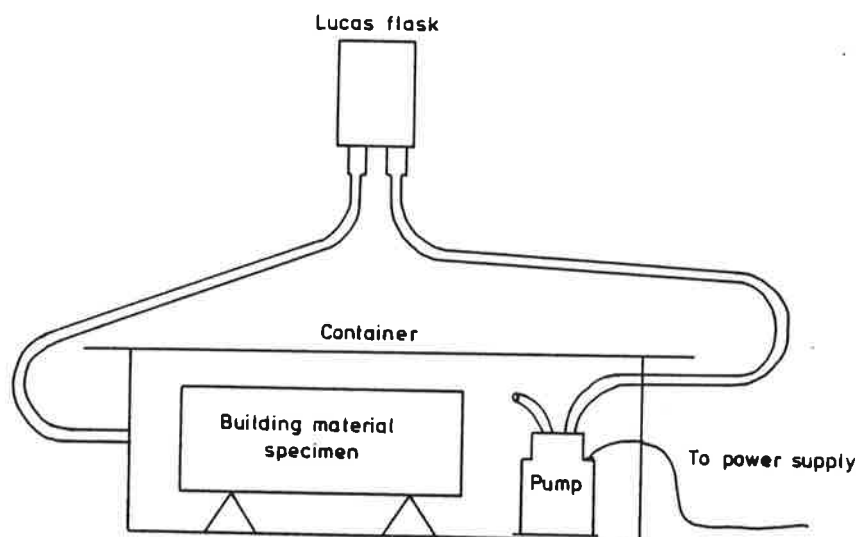


Figure B1 Container - Lucas flask arrangement for radon emanation studies from building materials

around these leads. At opposing ends of the container are inlet and exit tubes connected to self-closing vacuum connectors. The lid of the container is screwed firmly to the body, with an intervening gasket liberally coated in vacuum grease. Immediately the specimen is sealed in the container, the container is flushed through with air from a compressed air cylinder that has been stored for at least 5 weeks. Air from such a container is free of radon-222 as any that was present when the cylinder was filled will have decayed by the time of its use.

Approximately 10 h after the container is flushed with radon-free air a 160 ml Lucas scintillation flask is connected to the container by means of the vacuum connectors. The flask is also filled with aged air. The pump within the container is activated and air passes from the container through the scintillation flask and back to the container. The pump is operated long enough to ensure that the air in the container and the scintillation flask is of uniform radon-222 concentration. The pump is then stopped and the scintillation flask disconnected.

The scintillation flask comprises a cylindrical body, with a top-plate containing two self-closing vacuum connectors (female) and a transparent window at the bottom. The metallic inner surfaces of the cell are coated with zinc sulphide scintillation material. After admitting air containing radon-222 to the flask the short-lived decay products of radon-222 will be formed and after about 3 h they will be in equilibrium with the parent radon-222. At equilibrium there will be three alpha particles emitted within the flask for each radon-222 atom that decays. The flask is placed in an apparatus such that its transparent

window is in close proximity to the photocathode of a photomultiplier tube. The number of alpha-particles released within the flask for a given period can then be recorded. To achieve maximum sensitivity a long counting period, usually in excess of nine h, is used.

At further times of approximately 24, 36 and 48 h after the container is sealed and flushed with aged air, further samples of the enclosed air are taken with the Lucas flask. The same flask is used for each measurement as this obviates the need to make corrections for the dilution of the container air at the sampling time. When the Lucas flask is connected to the container, the pump within the container is operated to mix the air and circulate it through the flask. The flask is disconnected and placed in the counting apparatus. At the time of the sample the radon concentration in the container and in the flask are similar, and it would decay at the same rate if the container did not contain a source of radon. During the counting period, while the flask is not connected to the container, the radon concentration in the container rises more than it would if the flask were attached. As the flask volume is less than 10% of the free volume in the container no significant reduction in emanation rate is experienced. When the flask is connected to the container for the next sample, the concentration of radon in the flask and container will, essentially, be the same, as if the flask had been permanently connected to the container.

From the measurements outlined above, the concentration of radon-222 in the container air at the sampling times is obtained. For each measurement there is an uncertainty due to the random nature of radioactive decay. This uncertainty is often referred to as due to counting statistics and is always highest on the determination at the first sampling time because the lowest concentration is then being measured. As the concentrations measured are likely to be low it is essential to use a flask that has a low inherent background counting rate and that the background counting rate is determined over as long a period as is practical:

When a flask is counted the integrated count,  $C_T$ , (from  $T_1$  to  $T_2$  after the sample) is recorded and comprises two components

$$C_T = C_R + C_B \quad \dots(B1)$$

where  $C_R$  is the count due to radon in the flask

and  $C_B$  is the count due to the background count rate of the flask.

$$C_R = \frac{C_0}{\lambda_R} [\exp(-\lambda_R T_1) - \exp(-\lambda_R T_2)] \quad \dots(B2)$$

where  $\lambda_R$  is the decay constant of radon-222,  $0.00755 \text{ h}^{-1}$

$C_0$  the count rate ( $\text{h}^{-1}$ ) due to radon-222 in the flask at the sampling time ( $T = 0$ )

and  $B$  is the background count rate ( $\text{h}^{-1}$ ) of the flask and is determined over  $N$  h.

The total background count recorded in  $N$  hours is  $NB$  counts. Assuming  $NB$  to be a Poisson variate the standard deviation is  $(NB)^{\frac{1}{2}}$ . The standard deviation on the background count rate is then

$$(NB)^{\frac{1}{2}}/N = (B/N)^{\frac{1}{2}} \quad \text{.....(B3)}$$

Let

$$\frac{1}{\lambda_R} [\exp(-\lambda_R T_1) - \exp(-\lambda_R T_2)] = K \quad \text{.....(B4)}$$

Then

$$C_o = \frac{C_T}{K} - \frac{(T_2 - T_1)B}{K} \quad (\text{counts/h}^{-1}) \quad \text{.....(B5)}$$

The standard deviation on  $C_o$  (assuming Poisson statistics are valid) is then

$$S_o = \frac{1}{K} \left[ C_T - (T_2 - T_1)^2 \frac{B}{N} \right]^{\frac{1}{2}} \quad \text{.....(B6)}$$

For a typical low-activity air sample taken 10 h after sealing:

$$B = 20 \text{ h}^{-1}$$

$$N = 72 \text{ h}$$

$$T_1 = 2$$

$$T_2 = 13.5$$

$$C_T = 500.$$

Then  $C_o = 24.9 \text{ counts h}^{-1}$  and  $S_o = 2.14 \text{ counts h}^{-1}$ , or in this case a coefficient of variation of 8.5%. This is a typical result from a clay brick with a sample taken 10 h after sealing. The coefficient of variation improves as later samples are taken and the concentration being determined is higher. The count rate,  $C_o$ , is directly proportional to the radon-222 concentration in the flask at the time of taking the air sample.

Assuming the radon-222 emanation rate from the specimen into the container air is at a constant rate (which will be essentially true providing the specimen has not been sealed into the container for a period much in excess of the half-life of radon-222), then the count rate from the flask as a function of the sampling time,  $t$ , is

$$C_o = C_s [1 - \exp(-\lambda_R t)] \quad \text{.....(B7)}$$

$C_s$  is the steady-state, or equilibrium, count rate which  $C_o$  approaches asymptotically for large  $t$  ( $C_o = 0.995 C_s$  at  $t = 700 \text{ h}$ ).

Because of statistical uncertainties associated with counting disintegrations from radioactive material, which depend upon both the activity of the source and the period over which measurements take place, the measurements of radon-222 concentration taken during the measurement of a building material do not have equal coefficients of variation. The four values of radon concentration

are fitted to the function given in (B7) by a weighted least-squares regression to give

$$C_s = \frac{\sum_i \frac{C_{oi} [1 - \exp(-\lambda_R t_i)]}{S_{oi}^2}}{\sum_i \frac{[1 - 2\exp(-\lambda_R t_i) - \exp(-2\lambda_R t_i)]}{S_{oi}^2}} \quad \text{.....(B8)}$$

In equation (B8) the determination of  $C_o$  and  $S_o$  for sampling time  $t_i$  is represented by  $C_{oi}$  and  $S_{oi}$ .

A typical example for a specimen of clay brick is given in Table B1. For each sampling time,  $t$ , the count rate,  $C_o$ , and standard deviation  $S_o$  are calculated from equations (B2) and (B6), respectively. These are the values shown in columns five and six. The equilibrium counts  $h^{-1}$ ,  $C_s$ , is then found using the four individual measurements and substituting in equation (B8). In this case  $C_s$  is 442 counts  $h^{-1}$ . This value is then used in equation (B7) to produce the values in column seven of Table B1.

Table B1  
Growth of radon-222 concentration in a container containing  
a clay brick

Time, t, of air sample (h)	Flask counted		Gross count	Count rate at, t (counts $h^{-1}$ )		Co' count rate at t from equation (B7)
	From (h)	To (h)		$C_o$	$S_o$	
9.92	11.92	23.42	568	35.4	1.92	32.0
23.5	25.5	33.5	623	64.7	3.09	72.1
33.9	35.9	47.4	1273	100	3.11	100
44.7	46.7	51.2	653	133	5.77	127

The scintillation flask used in the above measurements had a background counting rate of 16 counts  $h^{-1}$  determined over 16 h.

The experimental results for two bricks of differing radon-222 emanation rates are shown in Figure B2. In Figure B2 the solid lines represent the theoretical growth curves given by equation (B7) and the experimental points are shown with error bars which represent one standard deviation due to counting statistics only.

As the counts recorded by the Lucas flask method are derived from the parent radon and its decay products the counts are not independent and simple Poisson statistics do not apply. In this case the variance of C observed counts is not

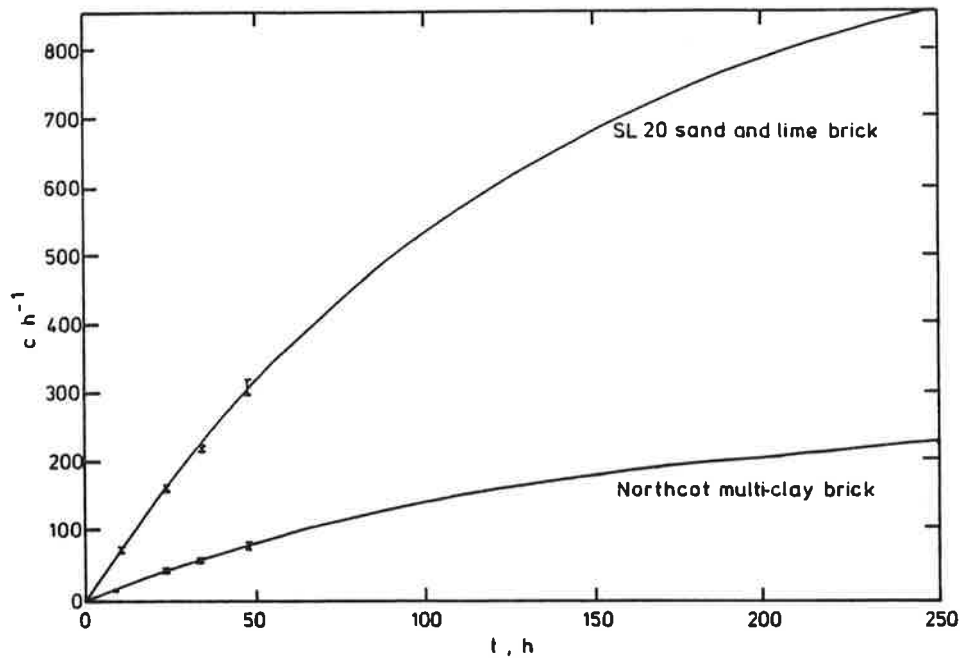


Figure B2 Growth curves for Northcot multi-clay brick and SL20 sand and lime brick

$C$ , but  $JC^{(2)}$ . Where  $J$  is a complex function of the efficiency of the Lucas flask, the time delay between filling the flask and starting the alpha count and the duration of the alpha count.

In equation (B6),  $C_T$  can be written as

$$C_T = C_T - C_B + C_B$$

where  $C_T - C_B$  is  $C_R$  from equation (B1).

The variance of  $C_R$  is  $JC_R$

$$JC_R = JC_T - JC_B$$

The variance of  $C_T$  is then

$$JC_T - JC_B + C_B$$

and this can be reinserted in (B6) to give

$$S_o = \frac{1}{K} \left[ (JC_T + (1-J)(T_2 - T_1)B + (T_2 - T_1)^2 \frac{B}{N}) \right]^{\frac{1}{2}} \quad \dots(B9)$$

For  $J = 1$ , equations (B6) and (B8) are identical, and in the application used here the disregarding of the  $J$  factor makes little difference to the results, as is shown in Table B2. This table reproduces the basic data of Table B1, but introduces the  $J$  factors. Each counting regime has been allotted a  $J$  value which, while not precise, is of the right order.

Table B2

Growth of radon-222 concentration in a container containing a  
clay brick taking account the appropriate J factors

Time, t, of air sample (h)	Flask counted		Gross count	J	Count rate at t		$C_0^1$
	From (h)	To (h)			$C_0$	$S_0$	
9.92	11.92	23.42	568	2.2	35.4	3.14	31.9
23.5	25.5	33.5	623	1.7	64.7	4.20	71.8
33.9	35.9	47.4	1273	2.7	100	5.26	99.8
44.7	46.7	51.2	653	1.4	133	6.91	127

As can be seen from the final columns of Tables B1 and B2, little difference is obtained when the more correct statistics are applied. In this case the value of 442 counts  $h^{-1}$  for  $C_S$  derived from Table B2 is the same (correct to three significant figures) as that from Table B1. Other examples have been investigated and the largest difference in  $C_S$  values between the two methods was just under 2%. These results are expected in this application as, by either method (with or without J), the smaller coefficients of variation are obtained for the measurements at the later times.

### 3. DETERMINATION OF THE CONCENTRATION OF THE DECAY PRODUCTS OF RADON BY AIR SAMPLING AND THE CALCUALTION OF RADON PRODUCTION RATES AND NORMALISED VALUES

#### 3.1 Determination of the concentration in air of the decay products of radon-222

A number of methods have been devised for assessing the concentration of the decay products of radon-222 in air by passing a sample of the air of interest through a filter paper and then analysing the decay of deposited nuclides on the filter paper after sampling has been completed. If the air contains the decay products of radon-222 only, or if the decay products of radon-220 present in air are at a very low concentration compared with those of radon-222, then three integrated gross alpha counts have to be taken after sampling to determine the concentrations of polonium-218, lead-214 and bismuth-214. A common procedure (devised by Thomas) is to take an air sample at constant flow rate through the filter paper from 0-5 minutes, and then carry out gross alpha counting of the material deposited on the filter paper for periods from 7-10, 11-26 and 27-35 minutes to give counts  $C_1$ ,  $C_2$  and  $C_3$ , respectively. The three counts obtained are substituted in three equations, one each for polonium-218, lead-214 and bismuth-214<sup>(3)</sup>. As the WL value in air is a linear combination of the decay product concentrations, an equation can also be found, that can be solved by the use of  $C_1$ ,  $C_2$  and  $C_3$ , to give the WL directly. If the only interest is in determining the WL value then any sample and counting regime will suffice, and adequate precision can be obtained by the use of one count only, provided a 40



minute delay, at least, is allowed<sup>(4)</sup> between the end of the sample and the start of the count.

If the individual decay product concentrations are required then the sampling time and the ensuing counting periods have to be chosen with care. However, with all sampling schemes where gross alpha counting takes place only after the termination of sampling, the uncertainty in the determination of polonium-218 concentration is large. This is due to the short half-life of polonium-218 (3.05 minutes), and in the case of Thomas's method 62% of the polonium-218 deposited on the filter paper will have decayed before the first gross alpha count is started.

To overcome this problem, an instrument was designed that permitted gross alpha counting as sampling was taking place, as well as for periods after sampling had ceased<sup>(5)</sup>. This instrument was originally designed for use in mines, using a model of the mine atmosphere that permitted only two counts to be taken. The first count to take place during sampling and the second count for a similar period after sampling. From the ratio of the counts, computed tables can be consulted which provide factors relating the first count to the polonium-218 concentration and the second count to the WL. This instrument and later versions of it were used to determine the concentration of the decay products of radon-222 in the general environment. Instead of relying on a model of the atmosphere the first count was taken during sampling, and two further counts were taken after sampling had ceased<sup>(6,7)</sup>. In general, the procedure is to count while sampling from 0 to  $T_1$  minutes. Sampling stops at  $T_1$  and then counts are made from  $T_1 + 1$  to  $T_2$  minutes and from  $T_2 + 1$  to  $T_3$  minutes, this can be denoted by I ( $T_1, T_2, T_3$ ). The most usual regime is I (15, 36, 57), ie sample and count from 0 to 15 minutes, count from 16 to 36 minutes and from 37 to 57 minutes to provide counts  $C_1, C_2$  and  $C_3$ , respectively. Table B3 demonstrates the advantages of counting while sampling in order to obtain a low uncertainty on the polonium-218 assessment. In this table the atmosphere considered contains 3.7

Table B3

Uncertainty in assessing polonium-218, lead-214, bismuth-214 concentrations and WL when the concentration in air is 3.7 kBq m<sup>-3</sup> each of polonium-218, lead-214 and bismuth-214, the sampling flow rate is 10 l min<sup>-1</sup> and the alpha counting efficiency is 0.2

	Thomas	Uncertainty (one SD) using I ( $T_1, T_2, T_3$ )		
		I (5, 25, 35)	I (10, 26, 35)	I (15, 36, 57)
Polonium-218	11.8%	5.0%	4.4%	3.0%
Lead-214	3.7%	3.2%	2.9%	1.0%
Bismuth-214	3.9%	2.8%	2.5%	1.4%
WL	1.4%	1.1%	1.0%	0.3%

kBq m<sup>-3</sup> each of polonium-218, lead-214 and bismuth-214. The sampling flow rate is 10 l min<sup>-1</sup> and the counting efficiency is 0.2. The uncertainty on the assessment of each parameter is given for the Thomas method, and for two procedures of I (T<sub>1</sub>, T<sub>2</sub>, T<sub>3</sub>) which have the same overall measurement time as the Thomas method (35 minutes): the first also has the same sampling time of 5 minutes. Finally the figures achievable with the currently used regime, I (15, 36, 57), are given.

In practice, a larger pump which permits sampling at about 40 l min<sup>-1</sup> is used for environmental monitoring and the uncertainties are further reduced, as the uncertainty is inversely proportional to the square root of the flow rate.

The latest versions of the instruments for measuring the concentration of radon decay products are fully automatic for the decay products of radon-222, and with the minimum of operator intervention will extend the number of alpha counts recorded can be extended from three to five or six, thus permitting the assessment of the concentrations of the decay products of radon-220 as well<sup>(8)</sup>.

### 3.2 Measurement of the radon-222 production rate

The measurement of polonium-218 concentration in indoor air and in the open air is used to estimate the rate of production of radon-222 in the room being studied.

In general,

$$\frac{dQ_R}{dt} = k - \lambda_R Q_R - j Q_R + j Q_R^1 \quad \text{.....(B10)}$$

where  $Q_R$  is the concentration (atoms m<sup>-3</sup>) of radon-222 in room air

$k$  the radon-222 production rate of the room in atoms m<sup>-3</sup> of room air h<sup>-1</sup>

$j$  is the ventilation rate (h<sup>-1</sup>) with air exchange solely between the room and the outside air

$Q_R^1$  be the concentration (atoms m<sup>-3</sup>) of radon-222 in outside air

and  $\lambda_R$  the decay constant of radon-222 (h<sup>-1</sup>).

In the steady state  $dQ_R/dt = 0$  and

$$k = Q_R (\lambda_R + j) - j Q_R^1 \quad \text{.....(B11)}$$

However, in all practical cases  $j \gg \lambda_R$  and

$$k = Q_R j - j Q_R^1 \quad \text{.....(B12)}$$

The radon-222 production rate could be determined from equation (B12), but in practice it is more convenient to measure the concentration of polonium-218 than that of radon-222.

For polonium-218 the corresponding equation to (B10) is

$$\frac{dQ_A}{dt} = \lambda_R Q_R - \lambda_A Q_A - j Q_A + j Q_A^1 \quad \text{.....(B13)}$$

where  $\lambda_A$  is the decay constant of polonium-218 ( $\text{h}^{-1}$ )  
 $Q_A$  the concentration (atoms  $\text{m}^{-3}$ ) of polonium-218 in room air  
and  $Q_A^1$  the concentration (atoms  $\text{m}^{-3}$ ) of polonium-218 in outside air.  
In the steady state  $dQ_A/dt = 0$ , and from (B12)

$$Q_R = \frac{k + jQ_R^1}{j}$$

Hence

$$\frac{\lambda_R k + j\lambda_R Q_R^1}{j} - Q_A (\lambda_A + j) + jQ_A^1 = 0$$

$$\lambda_R k = jQ_A (\lambda_A + j) - j^2 Q_A^1 - j\lambda_R Q$$

.....(B14)

In outside air close to the ground polonium-218 will be in equilibrium with radon-222 and

$$\lambda_R Q_R^1 = \lambda_A Q_A^1$$

$$\lambda_R k = jQ_A (\lambda_A + j) - j^2 Q_A^1 - j\lambda_A Q_A^1$$

$$\lambda_R k = \frac{jQ_A \lambda_A (\lambda_A + j)}{\lambda_A} + \frac{j^2 \lambda_A Q_A^1}{\lambda_A} - \frac{j\lambda_A \lambda_A Q_A^1}{\lambda_A}$$

.....(B15)

but\*  $\lambda_A Q_A$  is the concentration of polonium-218 in indoor air ( $\text{Bq m}^{-3}$ ),  $C_I$ ,  
and  $\lambda_A Q_A^1$  is the concentration of polonium-218 in outdoor air ( $\text{Bq m}^{-3}$ ),  $C_O$ .  
 $\lambda_R k$  is the radon-222 production rate  $K$  ( $\text{Bq m}^{-3} \text{h}^{-1}$ )

$$K = \frac{jC_I (\lambda_A + j)}{\lambda_A} - \frac{j^2 C_O}{\lambda_A} - jC_O$$

$$K = j (1 + j/\lambda_A) (C_I - C_O)$$

.....(B16)

or for  $\lambda_A = 13.64 \text{ h}^{-1}$

$$K = j (1 + 0.0733j) (C_I - C_O)$$

.....(B17)

Thus, by measuring the concentration of polonium-218 in indoor air and in the air outside, together with the ventilation rate, equation (B17) is used to effect an estimate of the radon production rate. From a knowledge of the radon-222 production rate the actual concentration of radon-222 and its decay products in room air at any given ventilation rate can be predicted. The above analysis assumes that the decay products of radon-222 are removed from room air solely by radioactive decay and ventilation to the outside air. In practice some decay products are removed from room air by plate-out to room surfaces. Plate-out can be taken into account in the above analysis by adding an additional loss term to

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\*As  $\lambda_R$  and  $\lambda_A$  are in units  $\text{h}^{-1}$  the true conversion from atoms  $\text{m}^{-3}$  to  $\text{Bq m}^{-3}$  should be  $(\lambda_A Q_A)/3600$ , but as the factor  $1/3600$  is a scaling factor appearing on both sides of equation (B-16) it has been omitted for clarity.

equation (B13) that accounts for plate-out removal of polonium-218 from room air.

Equation (B13) then becomes

$$\frac{dQ_A}{dt} = \lambda_R Q_R - \lambda_A Q_A - \lambda_P Q_A - jQ_A + jQ_A^1 \quad \text{.....(B18)}$$

where  $\lambda_P$  is the removal-rate constant for the plate-out mechanism.

Equation (B17) then becomes

$$K = j (1 + 0.0733j) (C_I - C_O) + 0.0733 j \lambda_P C_I \quad \text{.....(B19)}$$

Unfortunately  $\lambda_P$  is difficult to quantify, as plate-out removal constants are vastly different for the attached and unattached polonium-218, and the fraction unattached depends on the particular location being investigated. Methods of measuring plate-out rate constants are being investigated to enable representative values to be determined.

### 3.3 Normalised WL values

Let a room have a source term of  $k$  atoms  $m^{-3} s^{-1}$  of radon-222, then for radon-222

$$\frac{dQ_R}{dt} = k - \lambda_R Q_R - jQ_R + jQ_R^1$$

where  $\lambda_R$  is the decay constant of radon-222 ( $s^{-1}$ )

and  $j$  the ventilation rate ( $s^{-1}$ ).

In the steady state  $dQ_R/dt = 0$  and

$$Q_R (\lambda_R + j) = k + jQ_R^1$$

but for all practical ventilation rates  $j \gg \lambda_R$  and

$$Q_R = \frac{k}{j} + Q_R^1 \quad \text{.....(B20)}$$

For polonium-218

$$\frac{dQ_A}{dt} = \lambda_R Q_R - \lambda_A Q_A - jQ_A + jQ_A^1$$

where  $\lambda_A$  is the decay constant of polonium-218 ( $s^{-1}$ ).

In the steady state

$$Q_A (\lambda_A + j) = \frac{\lambda_R k}{j} + \lambda_R Q_R^1 + jQ_A^1$$

$$Q_A = \frac{\lambda_R k}{j(\lambda_A + j)} + \frac{\lambda_R Q_R^1}{(\lambda_A + j)} + \frac{jQ_A^1}{(\lambda_A + j)} \quad \text{.....(B21)}$$

For lead-214

$$\frac{dQ_B}{dt} = \lambda_A Q_A - \lambda_B Q_B - jQ_B + jQ_B^1$$

where  $Q_B$  is the concentration (atoms  $m^{-3}$ ) of lead-214 in room air

$\lambda_B$  the decay constant of lead-214 ( $s^{-1}$ )

and  $Q_B^1$  the concentration (atoms  $m^{-3}$ ) of lead-214 in outside air.

In the steady state

$$Q_B (\lambda_B + j) = \frac{\lambda_A \lambda_R k}{j(\lambda_A + j)} + \frac{\lambda_A \lambda_R Q_R^1}{(\lambda_A + j)} + \frac{j \lambda_A Q_A^1}{(\lambda_A + j)} + j Q_B^1$$

$$Q_B = \frac{\lambda_A \lambda_R k}{j(\lambda_A + j)(\lambda_B + j)} + \frac{\lambda_A \lambda_R Q_R^1}{(\lambda_A + j)(\lambda_B + j)} + \frac{j \lambda_A Q_A^1}{(\lambda_A + j)(\lambda_B + j)} + \frac{j Q_B^1}{(\lambda_B + j)} \dots\dots(B22)$$

For bismuth-214

$$\frac{dQ_C}{dt} = \lambda_B Q_B - \lambda_C Q_C - j Q_C + j Q_C^1$$

where  $Q_C$  is the concentration (atoms  $m^{-3}$ ) of bismuth-214 in room air

$\lambda_C$  the decay constant of bismuth-214 ( $s^{-1}$ )

and  $Q_C^1$  the concentrations (atoms  $m^{-3}$ ) of bismuth-214 in outside air.

In the steady state

$$Q_C (\lambda_C + j) = \frac{\lambda_A \lambda_B \lambda_R k}{j(\lambda_A + j)(\lambda_B + j)} + \frac{\lambda_A \lambda_B \lambda_R Q_R^1}{(\lambda_A + j)(\lambda_B + j)} + \frac{j \lambda_A \lambda_B Q_A^1}{(\lambda_A + j)(\lambda_B + j)}$$

$$+ \frac{j \lambda_B Q_B^1}{(\lambda_B + j)} + j Q_C^1$$

$$Q_C = \frac{\lambda_A \lambda_B \lambda_R k}{j(\lambda_A + j)(\lambda_B + j)(\lambda_C + j)} + \frac{j \lambda_B Q_B^1}{(\lambda_B + j)(\lambda_C + j)}$$

$$+ \frac{\lambda_A \lambda_B \lambda_R Q_R^1}{(\lambda_A + j)(\lambda_B + j)(\lambda_C + j)} + \frac{j \lambda_A \lambda_B Q_A^1}{(\lambda_A + j)(\lambda_B + j)(\lambda_C + j)} + \frac{j Q_C^1}{(\lambda_C + j)} \dots\dots(B23)$$

$\lambda_R k$  is the radon-222 source term  $S_0$  ( $Bq m^{-3} s^{-1}$ ) and the activity concentrations ( $Bq m^{-3}$ ) are given by

$$C_A = \lambda_A Q_A, C_B = \lambda_B Q_B \text{ and } C_C = \lambda_C Q_C$$

where  $C_A$  is the concentration ( $Bq m^{-3}$ ) of polonium-218 in room air

$C_B$  the concentration ( $Bq m^{-3}$ ) of lead-214 in room air

and  $C_C$  the concentration ( $Bq m^{-3}$ ) of bismuth-214 in room air.

Similarly, the activity concentrations in the open air are given by

$$C_A^1 = \lambda_A Q_A^1, C_B^1 = \lambda_B Q_B^1 \text{ and } C_C^1 = \lambda_C Q_C^1$$

where  $C_A^1$  is the concentration ( $Bq m^{-3}$ ) of polonium-218 in outside air

$C_B^1$  the concentration ( $Bq m^{-3}$ ) of lead-214 in outside air

and  $C_C^1$  the concentration ( $Bq m^{-3}$ ) of bismuth-214 in outside air.

From (B21)

$$C_A = \frac{\lambda_A S_0}{j(\lambda_A + j)} + \frac{\lambda_A C_R^1}{(\lambda_A + j)} + \frac{j C_A^1}{(\lambda_A + j)} \dots\dots(B24)$$

where  $C_R^1$  is the concentration ( $Bq m^{-3}$ ) of radon-22 in outside air.

From (B22)

$$C_B = \frac{\lambda_A \lambda_B S_0}{j(\lambda_A + j)(\lambda_B + j)} + \frac{\lambda_A \lambda_B C_R^1}{(\lambda_A + j)(\lambda_B + j)} + \frac{j \lambda_B C_A^1}{(\lambda_A + j)(\lambda_B + j)} + \frac{j C_B^1}{(\lambda_B + j)} \dots\dots(B25)$$

From (B23)

$$C_C = \frac{\lambda_A \lambda_B \lambda_C S_0}{j(\lambda_A + j)(\lambda_B + j)(\lambda_C + j)} + \frac{\lambda_A \lambda_B \lambda_C C_R^1}{(\lambda_A + j)(\lambda_B + j)(\lambda_C + j)} + \frac{j \lambda_B \lambda_C C_A^1}{(\lambda_A + j)(\lambda_B + j)(\lambda_C + j)} + \frac{j \lambda_C C_B^1}{(\lambda_B + j)(\lambda_C + j)} + \frac{j C_C^1}{(\lambda_C + j)} \dots\dots(B26)$$

If in the open air the decay products of radon-222 are in equilibrium with the parent, then

$$C_R^1 = C_A^1 = C_B^1 = C_C^1$$

$$C_A = \frac{\lambda_A S_0}{j(\lambda_A + j)} + \frac{\lambda_A C_A^1}{(\lambda_A + j)} + \frac{j C_A^1}{(\lambda_A + j)}$$

$$C_A = \frac{\lambda_A S_0}{j(\lambda_A + j)} + C_A^1 \dots\dots(B27)$$

$$C_B = \frac{\lambda_A \lambda_B S_0}{j(\lambda_A + j)(\lambda_B + j)} + \frac{\lambda_A \lambda_B C_B^1}{(\lambda_A + j)(\lambda_B + j)} + \frac{j \lambda_B C_B^1}{(\lambda_A + j)(\lambda_B + j)} + \frac{j C_B^1}{(\lambda_B + j)}$$

$$C_B = \frac{\lambda_A \lambda_B S_0}{j(\lambda_A + j)(\lambda_B + j)} + C_B^1 \dots\dots(B28)$$

$$C_C = \frac{\lambda_A \lambda_B \lambda_C S_0}{j(\lambda_A + j)(\lambda_B + j)(\lambda_C + j)} + \frac{\lambda_A \lambda_B \lambda_C C_C^1}{(\lambda_A + j)(\lambda_B + j)(\lambda_C + j)} + \frac{j \lambda_B \lambda_C C_C^1}{(\lambda_A + j)(\lambda_B + j)(\lambda_C + j)} + \frac{j \lambda_A \lambda_C C_C^1}{(\lambda_B + j)(\lambda_C + j)} + \frac{j C_C^1}{(\lambda_C + j)}$$

$$C_C = \frac{\lambda_A \lambda_B \lambda_C S_0}{j(\lambda_A + j)(\lambda_B + j)(\lambda_C + j)} + C_C^1 \dots\dots(B29)$$

The working level (WL) is a linear combination of the activity concentrations of polonium-218, lead-214 and bismuth-214 weighted according to the potential energy contribution per unit activity concentration of each nuclide

$$WL = K_A C_A + K_B C_B + K_C C_C \dots\dots(B30)$$

where  $K_A$ ,  $K_B$  and  $K_C$  are the weighting factors.

Thus

$$\begin{aligned}
 WL &= \frac{K_A \lambda_A S_0}{j(\lambda_A + j)} + K_A C_A^1 + \frac{K_B \lambda_A \lambda_B S_0}{j(\lambda_A + j)(\lambda_B + j)} + K_B C_B^1 \\
 &+ \frac{K_C \lambda_A \lambda_B \lambda_C S_0}{j(\lambda_A + j)(\lambda_B + j)(\lambda_C + j)} + K_C C_C^1 \\
 WL &= S_0 \left[ \frac{K_A \lambda_A}{j(\lambda_A + j)} + \frac{K_B \lambda_A \lambda_B}{j(\lambda_A + j)(\lambda_B + j)} + \frac{K_C \lambda_A \lambda_B \lambda_C}{j(\lambda_A + j)(\lambda_B + j)(\lambda_C + j)} \right] \\
 &+ K_A C_A^1 + K_B C_B^1 + K_C C_C^1 \quad \dots(B31)
 \end{aligned}$$

However,  $K_A C_A^1 + K_B C_B^1 + K_C C_C^1$  is simply the WL value in the open air,  $WL_0$ .

The above derivation has been performed using seconds as the unit of time. This preserves the strict relationship for the conversion from atoms  $m^{-3}$  to Bq  $m^{-3}$  ( $C = \lambda Q$ ), but any consistent set of units may be used and it is more usual to have ventilation rate in units  $h^{-1}$ , where the following can be substituted in equation (B31).

$$\begin{aligned}
 K_A &= 2.786 \cdot 10^{-5} \text{ WL (Bq } m^{-3})^{-1} \text{ for polonium-218} \\
 K_B &= 1.370 \cdot 10^{-4} \text{ WL (Bq } m^{-3})^{-1} \text{ for lead-214} \\
 K_C &= 1.008 \cdot 10^{-4} \text{ WL (Bq } m^{-3})^{-1} \text{ for bismuth-214} \\
 \lambda_A &= 13.64 \text{ h}^{-1} \\
 \lambda_B &= 1.551 \text{ h}^{-1} \\
 \lambda_C &= 2.111 \text{ h}^{-1}
 \end{aligned}$$

to give, with  $S_I$  now the radon-222 source term in Bq  $m^{-3} h^{-1}$ ,

$$\begin{aligned}
 WL &= 10^{-3} S_I \left[ \frac{0.3800}{j(13.64+j)} + \frac{2.899}{j(13.64+j)(1.551+j)} \right. \\
 &\left. + \frac{4.502}{j(13.64+j)(1.551+j)(2.111+j)} \right] + WL_0 \quad \dots(B32)
 \end{aligned}$$

It is possible to normalise a WL value in indoor air measured at ventilation rate  $j(h^{-1})$  to that which would occur at some other ventilation rate. For convenience, normalisation to the value occurring at one air change per hour is often used. The value of WL at one air change per hour,  $WL_N$ , is found by simply substituting 1.0 for  $j$  in equation (B32).

This yields

$$WL_N = 1.423 \cdot 10^{-4} S_I + WL_0 \quad \dots(B33)$$

Then from equation (B32) and (B33)

$$WL_N = [WL - WL_0] \left[ \frac{j(13.64+j)(1.551+j)(2.111+j)}{2.670(1.551+j)(2.111+j) + 20.37(2.111+j) + 31.64} \right] + WL_0 \quad \dots(B34)$$

#### 4. PASSIVE RADON DETECTORS

Many dielectric materials suffer sub-microscopic damage when struck by alpha-particles or other heavy charged particles. Suitable treatment can enlarge the damage tracks to a size whereby they may be seen under a lower power microscope. A variety of such materials have been used to measure the concentration of radon-222 in air. Some materials that have been used for this purpose are polycarbonate<sup>(9)</sup>, forms of cellulose nitrate, such as LR-115 and CA-80-15<sup>(10)</sup>, and allyl diglycol carbonate (CR-39), which is the material used in the measurements reported here<sup>(11)</sup>.

When an alpha particle impinges upon a thin sheet of CR-39 the alpha particle penetrates a short distance before being stopped. Chemical bonds along the path of the alpha particle in the dielectric are disrupted but there is no visible evidence of this. If the dielectric is etched in a caustic solution the material damaged by the alpha particle etches at a faster rate than the bulk of the material. This procedure can increase the pit size such that it may be seen under a high-power microscope. If the etching is continued, in the presence of a high alternating electric field, the etched pits are broken down by the field to produce large tracks visible under a low-power microscope, or they can be seen on the display of a microfiche reader.

The passive radon detector currently used consists of a small plastic cup in the bottom of which is placed an 18.5 mm square of CR-39. A tubular spacer is then placed on top of this element and a second element placed on top of the spacer. A lid is then fitted to close the detector package. The lid prevents the ingress of dust and the decay products of radon-222 in the surrounding air. Radon-222 does diffuse into the container with a diffusion half-time of about 2 h. This half-time is short compared with the total measurement period and does not affect the integrated radon concentration, because when the exposure ceases the radon within the cup will take as long to diffuse out. The radon within the cup will form some decay products and the CR-39 will be struck by alpha particles from the radon itself and from those decay products formed in the volume of the cup. At the end of the exposure period the number of damage tracks per unit area of plastic is proportional to the average radon-222 concentration.

The processing of the detector involves a chemical etching stage of 1 h in 20% NaOH at 80°C, followed by electrochemical etching in 30% KOH with an applied field of 18.3 kV cm<sup>-1</sup> rms at 2 KHz at a temperature of 30°C for 6 h. Different sheets of CR-39 vary in sensitivity from 1.9 10<sup>-5</sup> to 4.3 10<sup>-5</sup> tracks mm<sup>-2</sup> for an exposure of 1 Bq m<sup>-3</sup> h, and the background track density varies from 0.15 to 0.40 tracks mm<sup>-2</sup>. Thus, for each sheet of CR-39, some detectors are used for calibration purposes and others to determine the background, and these figures are used in calculating the exposures received by the remaining detectors made from that sheet.

The electrochemical etching procedure results in large tracks that can



easily be counted and leads to a short processing time for the detector. Unfortunately, during the etching process it is possible for a track to be etched completely through the detector element, resulting in a short-circuit in the etching cell which terminates the etching of that element. It is not then possible to continue the processing of that element and that result is lost. This is the main reason for the inclusion of a second detecting element in each package. After the return of the dosimeter, the reserve element is kept in a radon-free atmosphere, and if its partner fails this reserve element can be chemically etched and assessed using a high-power microscope. A new method of assessing the passive radon detectors is being introduced. In this method, chemical etching alone is carried out and a fine scintillator material in an oil suspension is brushed on to the etched surface. The surplus oil is removed and the plastic interposed between an alpha-particle source and a photomultiplier tube. The scintillator granules trapped in the etched pits respond to the alpha particles and emit light. In this way the light output is proportional to the track density and a detector element can be read out in 5 s<sup>(12)</sup>.

##### 5. SIMPLE MODEL FOR COMPARING WHOLE-HOUSE AND SINGLE-ROOM RADON CONCENTRATIONS

As was stated in Section 5.2.4 of this report, the results of measurements of the radon-222 production rates in single rooms and under whole-house conditions produced quite different calculated mean radon concentrations under given ventilation rates in any given room. The following simple model, while not completely explaining the discrepancy, does demonstrate that concentrations in individual rooms may be quite different from that under whole-house conditions.

Figure B3 is a simple representation of a three compartment dwelling. A represents a ground-floor room, B a first-floor bedroom, and C is intended to

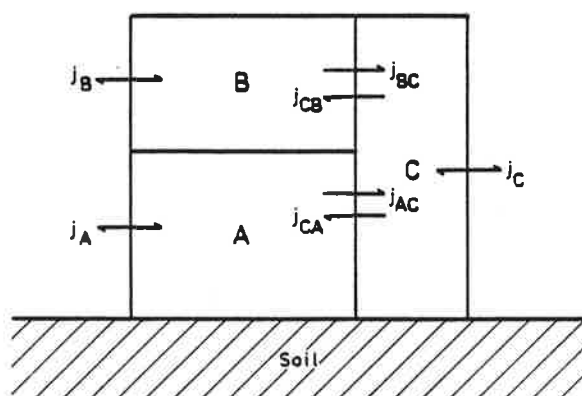


Figure B3 Simple model for comparing whole house and single room radon studies

represent the downstairs hall, stairway and upper landing. As shown, internal doors are assumed closed and for each room the air exchange rates are ( $h^{-1}$ ):

$j_A$  with the outside,  $j_{AC}$  with C (for room A);

$j_B$  with the outside,  $j_{BC}$  with C (for room B);

$j_C$  with the outside,  $j_{CA}$  with A and  $j_{CB}$  with B (for room C).

For simplicity the structure has no windows. The ceilings of all rooms emit no radon nor does the floor of B. It is also assumed that there is no direct path for radon transport from A to B. Let the radon production rates of rooms A, B and C be  $K_A$ ,  $K_B$  and  $K_C$   $Bq\ m^{-3}\ h^{-1}$ , respectively.

With all internal doors closed, and in the steady state, the concentration of radon-222 in rooms A, B and C, respectively, will be  $Q_A$ ,  $Q_B$  and  $Q_C$   $Bq\ m^{-3}$ , as for all practical air exchange rates the loss of radon-222 by decay is negligible.

$$Q_A = \frac{K_A + j_{CA}Q_C}{j_A + j_{AC}} \quad \text{.....(B35)}$$

$$Q_B = \frac{K_B + j_{CB}Q_C}{j_B + j_{BC}} \quad \text{.....(B36)}$$

$$Q_C = \frac{K_C + \frac{K_A j_{AC}}{j_A + j_{AC}} + \frac{K_B j_{BC}}{j_B + j_{BC}}}{j_C + j_{CB} + j_{CA} - \frac{j_{AC}j_{CA}}{j_A + j_{AC}} - \frac{j_{BC}j_{CB}}{j_B + j_{BC}}} \quad \text{.....(B37)}$$

If the internal doors are opened, but the ventilation rate to the outside air remains the same, there will be a uniform radon-222 concentration throughout the house,  $Q_H$ , where

$$Q_H = \frac{K_A j_{CA} j_{BC} + K_B j_{CB} j_{AC} + K_C j_{AC} j_{BC}}{j_A j_{CA} j_{BC} + j_B j_{AC} j_{CB} + j_C j_{AC} j_{BC}} \quad \text{.....(B38)}$$

Let the volumes of rooms A and B be equal and the volume of room C to be 1.5 that of A or B. Assume that the ground is the dominant source of radon-222 within the structure and that for room A the floor and ground contribute 2/3 of the total radon input and the walls 1/3, and, for simplicity, assume that the radon concentration in outdoor air is zero. Expect room C to have the highest ventilation rate, as the hall may contain a loose fitting exterior door and letter box, and choose the following values as reasonable examples:

$$\begin{aligned} K_A &= 21\ Bq\ m^{-3}\ h^{-1} & j_A &= 0.5\ h^{-1} & j_{AC} &= 0.15\ h^{-1} \\ K_B &= 7\ Bq\ m^{-3}\ h^{-1} & j_B &= 0.3\ h^{-1} & j_{CA} &= 0.1\ h^{-1} \\ K_C &= 22\ Bq\ m^{-3}\ h^{-1} & j_C &= 2.0\ h^{-1} & j_{BC} &= 0.15\ h^{-1} \\ & & & & j_{CB} &= 0.1\ h^{-1} \end{aligned}$$

Then, with the internal doors closed, the radon-222 concentration in room A will be  $34.4\ Bq\ m^{-3}$ ; in room B,  $18.6\ Bq\ m^{-3}$ ; in room C,  $13.6\ Bq\ m^{-3}$ . With the internal doors open the uniform radon-222 concentration throughout the house would be

16.1 Bq m<sup>-3</sup>. On the basis of a whole-house measurement the correct whole-house radon production rate would have been calculated as 17.3 Bq m<sup>-3</sup> h<sup>-1</sup>, and this would result in the prediction of a radon-222 concentration of 17.3 Bq m<sup>-3</sup> at a ventilation rate of one air change an hour. On the other hand, had a single-room measurement been carried out in room A the measured ventilation rate would have been 0.5 + 0.15 = 0.65 h<sup>-1</sup>, and the measured radon concentration of 34.4 Bq m<sup>-3</sup> would give a radon production rate of 22.4 Bq m<sup>-3</sup> h<sup>-1</sup>. This value is higher than the true value because the air exchange between rooms A and C has been neglected, it being assumed that the air exchange rate of 0.65 h<sup>-1</sup> is solely with the outside.

The single-room measurement would predict a radon-222 concentration in room A of 22.4 Bq m<sup>-3</sup> at a ventilation rate of one air change per hour, which is closer to the true value of 21 Bq m<sup>-3</sup> than is the 17.3 Bq m<sup>-3</sup> predicted from the whole house measurement.

Whole-house measurements may not, therefore, produce a figure for the radon production rate that permits a realistic prediction of the likely radon concentration in any particular room. Single-room measurements, while being prone to error due to air exchange between adjoining rooms, are more likely to produce estimated concentrations close to their true values. The lower the air exchange rate between rooms the closer the agreement will be.

#### 6. RADON DIFFUSION THROUGH MATERIALS

Radon will diffuse through many plastics as well as through porous media, such as soils and concrete. Figure B4 illustrates the case where radon is diffusing through a homogeneous material which contains no radium-226.

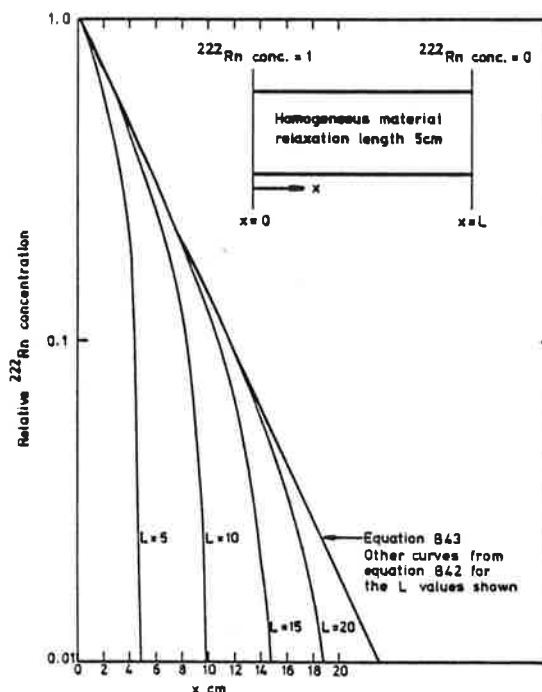


Figure B4 <sup>222</sup>Rn concentration profile in a homogeneous medium as function to length

One dimensional diffusion is characterised by two general equations. The first, from Fick's law, is  $J = -D(dC/dx)$

where  $J$  is the flux of activity across unit area of the material in unit time ( $\text{Bq m}^{-2} \text{ s}^{-1}$ )

$D$  the diffusion coefficient of radon in the material ( $\text{m}^2 \text{ s}^{-1}$ )

$C$  the concentration ( $\text{Bq m}^{-3}$ )

$x$  the direction of diffusion

and  $dC/dx$  the concentration gradient.

The second is

$$D \frac{d^2C}{dx^2} - \lambda C + K = 0 \quad \text{.....(B39b)}$$

where  $\lambda$  is the decay constant of radon-222 ( $2.1 \times 10^{-6} \text{ s}^{-1}$ )

and  $K$  is a term that only exists if the material contains a uniform distribution of radium-226 which produces radon-222 in the medium at a rate of  $K \text{ Bq m}^{-3} \text{ s}^{-1}$ .

The basic diffusion equation for a stable gas under steady-state conditions is

$$D \frac{d^2C}{dx^2} = 0$$

That is when the concentration of gas at any point in the system is constant with time.

The loss of a radioactive gas in transit through the medium due to radioactive decay is represented by  $\lambda C$ . For the system shown in Figure B4 the equation is simply

$$D \frac{d^2C}{dx^2} - \lambda C = 0 \quad \text{.....(B40)}$$

With boundary conditions  $C = C_0$  at  $x = 0$  and  $C = 0$  at  $x = L$  the solution is

$$C(x) = \frac{C_0 \sinh [(\lambda/D)^{1/2} (L - x)]}{\sinh [(\lambda/D)^{1/2} L]}$$

$(D/\lambda)^{1/2}$  has the dimensions of length and is often referred to as the relaxation length or diffusion length and given the symbol  $\ell_s$ .

For  $L > 3 \ell_s$ , and providing  $x$  is less than  $0.8L$ , equation (B41) reduces to the simpler form

$$C(x) = C_0 \exp [-(x/\ell_s)] \quad \text{.....(B42)}$$

From (B42) it can be seen that if this equation is valid then an increase in path length through the medium of  $\ell_s$  will result in the concentration being reduced by a factor of  $1/e$  (ie, the concentration falls by 63%). This, however, is only true for the conditions under which equation (B42) is valid and only

within a homogeneous medium. This reasoning cannot be applied when considering radon passing from soil into concrete or concrete into plastic, etc.

In porous media, such as soil, the concept of effective diffusion coefficient has to be introduced. This accounts for the situation that when radon diffuses into a reference area of porous medium the actual area participating in the diffusion process is predominantly the open pore areas, which form a small proportion of the total area. Culot et al.<sup>(13)</sup> have shown that the effective diffusion coefficient incorporates the porosity of the medium, as was shown in Section 6 of this report, with the effective diffusion coefficient denoted by  $K_e$  and the porosity by  $P$ .

Figure B5(a) depicts soil containing a uniform radium-226 concentration which produces radon-222 in the soil at a rate of  $K \text{ Bq m}^{-3} \text{ s}^{-1}$ . It is assumed that the radon concentration at the soil-air interface is maintained at zero.

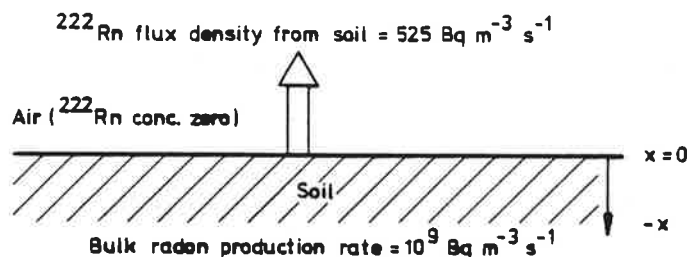


Figure B5(a)  $^{222}\text{Rn}$  emanation from soil

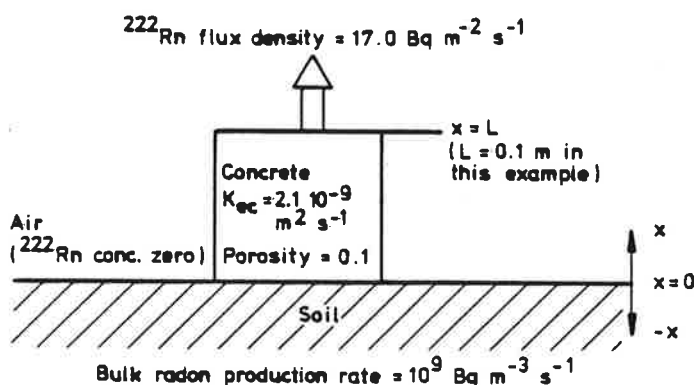


Figure B5(b)  $^{222}\text{Rn}$  emanation from the top of a concrete slab placed on the soil of Figure B5(a)

The relevant equation in this case is

$$\frac{K_{es}}{P_s} \frac{d^2 C(x)}{dx^2} - \lambda C(x) + K = 0 \quad \text{.....(B43)}$$

where  $K_{es}$  is the effective diffusion coefficient in soil  
and  $P_s$  the porosity of soil.

With the boundary conditions that at  $x = -\infty$ ,  $C(x) = K/\lambda$  and at  $x = 0$ ,  $C(x) = 0$ , this yields the solution

$$C(x) = \frac{K}{\lambda} \{1 - \exp[-x (\lambda P_s / K_{es})^{1/2}]\} \quad \text{.....(B44)}$$

or if  $[K_{es} / (\lambda P_s)]^{1/2} = \lambda_s$  (the relaxation length in soil)

$$C(x) = \frac{K}{\lambda} [1 - \exp(x/\lambda_s)] \quad \text{.....(B45)}$$

The flux at the air - soil interface is then given by

$$\begin{aligned} J &= -K_{es} \left. \frac{dC(x)}{dx} \right|_{x=0} \\ &= \frac{K}{\lambda} \frac{K_{es}}{\lambda_s} \end{aligned} \quad \text{.....(B46)}$$

For illustration purposes, with  $K/\lambda = 10^9$  Bq m<sup>-3</sup> and a typical value for  $K_{es}$  of  $5.25 \cdot 10^{-7}$  m<sup>2</sup> s<sup>-1</sup> (14) and  $P_s$  of 0.25, then  $\lambda_s = 1$  m.

The flux from the soil is then 525 Bq m<sup>-2</sup> s<sup>-1</sup>.

Figure B5(b) illustrates the case where a concrete slab of thickness  $L$  is placed on the soil of Figure B5(a). The concrete in this example is considered to be devoid of radium-226. In this case the relevant equations are

$$\frac{K_{es}}{P_s} \frac{d^2 C(x)}{dx^2} - \lambda C(x) + K = 0, \quad x \leq 0 \quad \text{.....(B47)}$$

$$\frac{K_{ec}}{P_c} \frac{d^2 C(x)}{dx^2} - \lambda C(x) = 0, \quad 0 \leq x \leq L \quad \text{.....(B48)}$$

where  $K_{ec}$  is the effective diffusion coefficient in concrete  
and  $P_c$  the porosity of concrete.

The boundary conditions are: at  $x = L$ ,  $C(x) = 0$ ; at  $x = 0$ ,  $C(x - Sx) = C(x + Sx)$   $Sx \rightarrow 0$ , ie, the concentration at the soil surface is equal to the concentration at the concrete base

$$K_{es} \left. \frac{dC(x)}{dx} \right|_{x=0} = K_{cs} \left. \frac{dC(x)}{dx} \right|_{x=0} \quad \text{flux continuity}$$

and at  $x = -\infty$ ,  $C(x) = K/\lambda$ .

The solution of equations (B47) and (B48) under these conditions yields the radon concentration in the soil to be given by ( $x \leq 0$ )

$$C(x) = \frac{K}{\lambda} \left[ 1 - \frac{\exp(x/\lambda_s)}{1 + (\lambda_c / K_{ec}) (K_{es} / \lambda_s) \tanh(L/\lambda_s)} \right] \quad \text{.....(B49)}$$

and the radon concentration in the concrete slab is given by, for  $0 \leq x \leq L$ ,

$$C(x) = \frac{K}{\lambda} \frac{\lambda_c}{K_{ec}} \frac{K_{es}}{\lambda_s} \left[ \frac{\cosh(x/\lambda_c) \tanh(L/\lambda_c) - \sinh(x/\lambda_c)}{1 + (\lambda_c/K_{ec}) (K_s/\lambda_s) \tanh(L/\lambda_c)} \right] \quad \text{.....(B50)}$$

where  $\lambda_c = (K_{ec}/\lambda P_c)^{1/2}$  is the relaxation length in concrete.

The flux through the top surface of the concrete slab is then

$$J_C = -K_{ec} \frac{dC(x)}{dx} \Big|_{x=L} = \frac{K}{\lambda} \frac{K_{es}}{\lambda_s} \left[ \cosh(L/\lambda_s) + (\lambda_c/K_{ec}) \sinh(L/\lambda_c) \right]^{-1} \quad \text{.....(B51)}$$

A typical value for the effective diffusion coefficient of concrete,  $K_{ec}$ , is

$2.1 \cdot 10^{-9} \text{ m}^2 \text{ s}^{-1}$  and a porosity,  $P_c$ , of 0.1 yields a relaxation length,  $\lambda_c$ , of 0.1 m.

Thus, considering a concrete slab 0.1 m thick laid on the soil, the flux through the top surface of the concrete is  $17.0 \text{ Bq m}^{-2} \text{ s}^{-1}$ , and the concrete reduces the flux from the soil by nearly 97% rather than the 63% that would be suggested by the relaxation length concept.

Such a reduction in flux is unlikely to persist with an actual concrete slab as cracking will occur, which facilitates the transport of radon through the slab. The above analysis can be extended to consider the effect of inserting a membrane in the concrete to act as a radon barrier. Figure B6 shows the relative flux from bare soil and through 0.1 m concrete slabs, with and without a  $2 \cdot 10^{-4} \text{ m}$  thick membrane of various diffusion coefficients. For a membrane of diffusion coefficient  $< 10^{-13} \text{ m}^2 \text{ s}^{-1}$ , extensive cracking of the concrete on either side of the membrane has a negligible effect on the emergent flux from the top of the slab.

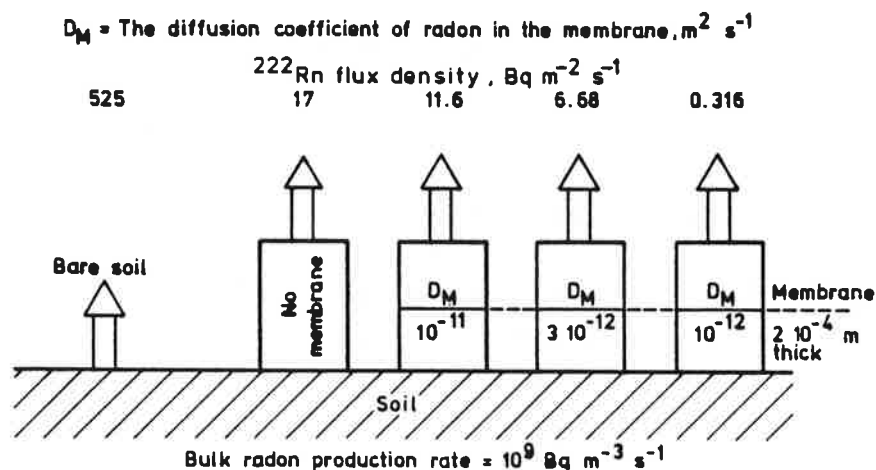


Figure B6  $^{222}\text{Rn}$  flux density through 0.1m concrete blocks with and without membranes

To determine the diffusion coefficient of radon-222 in a variety of membrane materials the apparatus, shown schematically in Figure B7, was constructed. Essentially, this consists of two cylindrical compartments. The two compartments can be bolted together, and around the flange through which the bolts pass are two O-rings. Each compartment has two hoses terminated by self closing vacuum connectors that can mate with a Lucas flask. The membrane material is situated between the compartments and held by the flange holding the two compartments together. Initially compartments A and B are filled with radon-free air. Container A is then filled with a high concentration of radon-222 and this concentration measured. It was intended that the concentrations in A and B would later be measured and the diffusion coefficient would be determined from the decrease in concentration in A and from the increase in concentration in B. Any discrepancy between the value of the diffusion coefficient calculated by these routes would be indicative of a leak in the system. The analysis of this system has proved more difficult than was first thought and is not complete. To date, the apparatus has been used in the simple mode of filling A with a high concentration of radon and maintaining the radon concentration in B close to

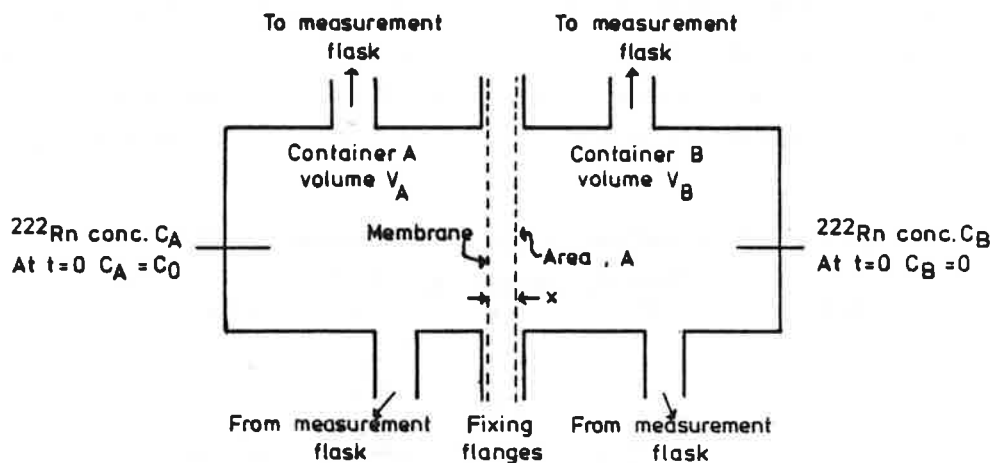


Figure B7 Measurements of the diffusion coefficient of radon in membrane materials



zero. The concentration of radon in container A is measured at a number of times after being filled with radon, and the concentration,  $C$ , at time  $t$  is given by

$$C_t = C_0 \exp \left[ \frac{-DA t}{V \lambda \tanh(x/\lambda)} \right] \exp(-\lambda t) \quad \text{.....(B52)}$$

where  $C_0$  is the radon concentration at  $t = 0$

$\lambda$  the decay concentration of radon-222 ( $2.1 \cdot 10^{-6} \text{ s}^{-1}$ )

$A$  the area of the membrane ( $\text{m}^2$ )

$V$  the volume of container A ( $\text{m}^3$ )

$x$  the thickness of the membrane ( $\text{m}$ )

$D$  the diffusion coefficient ( $\text{m}^2 \text{ s}^{-1}$ )

and  $\lambda = (D/\lambda)^{1/2}$  is the relaxation length  $m$ .

Equation (B52) is difficult to solve for  $D$ , but if  $x/\lambda < 0.1$ ,  $\tanh(x/\lambda) \approx x/\lambda$  and  $D$  is given by

$$D = \frac{Vx}{A} \left[ \frac{\ln(C_0/C_t)}{t} \right] - \lambda \quad \text{.....(B53)}$$

Equation (B53) is valid for a  $2 \cdot 10^{-4} \text{ m}$  thick membrane for  $D > 8.4 \cdot 10^{-12} \text{ m}^2 \text{ s}^{-1}$ . Equation (B53) also simplifies for  $x/\lambda > 4$ , as here  $\tanh(x/\lambda) \approx 1$ . However, for such low values of  $D$  ( $D < 5 \cdot 10^{-15} \text{ m}^2 \text{ s}^{-1}$ ) it would not be possible to measure the diffusion coefficient of a radioactive gas such as radon with a decay constant of  $2.1 \cdot 10^{-6} \text{ s}^{-1}$ . In this case  $DA/V\lambda$  for an apparatus of practical dimensions will be less than  $10^{-8} \text{ s}^{-1}$ , and such a small change in the apparent decay constant would not be detectable. For  $D < 8.4 \cdot 10^{-12} \text{ m}^2 \text{ s}^{-1}$  an iterative method must be used to solve equation (B52).

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## Appendix C

### Detailed results of the studies of radon-222 emanation from building materials

In Section 4.2 of this report, broad classes of radon-222 emanation rates were given for a number of materials. For example, all clay bricks were grouped together although there were 18 different types of clay brick tested. In this appendix the results for all sample types are given in Table C1 so that variations between types can be identified.

A number of manufacturers supplied details of the constituents of their products and the method of manufacture. These details are given in Table C2.

**Table C1**  
**Radon-222 emanation rates from building materials**

Sample	No.	Radon-222 emanation rates					
		$\text{Bq m}^{-3} \text{ s}^{-1} \times 10^{-3}$		$\text{Bq m}^{-2} \text{ s}^{-1} \times 10^{-6}$		$\text{Bq kg}^{-1} \text{ s}^{-1} \times 10^{-6}$	
		mean	range	mean	range	mean	range
Aerated concrete block	1	15	-	330	-	18	-
Insulating PFA block	1	4.9	-	80	-	3.6	-
Concrete block containing mine waste from Cornwall	1	30	-	530	-	16	-
Sintered colliery waste block	1	1.9	-	23	-	1.6	-
Concrete using granite from Gunislake Quarry	1	8.2	-	180	-	4.9	-
Thermolite Block	1	13	-	200	-	18	-
6" concrete block	3	2.5	2.3-2.7	90	85-97	1.3	1.3-1.4
4" concrete block	3	7.3	6.8-7.6	220	200-230	3.7	3.5-3.8
3" concrete block	3	3.0	1.9-3.9	77	50-100	1.7	1.2-2.2
Portion of paving slab	2	4.3	3.4-5.1	68	58-78	2.1	1.8-2.4
Light-weight concrete block	3	4.1	2.3-5.1	120	68-150	4.7	2.6-5.7
Aerated concrete block	1	5.5	-	99	-	6.8	-
Brick composed of colliery waste	1	0.41	-	5.6	-	0.24	-
Brick made from oil shale	1	17	-	290	-	11	-
Motherwell colliery shale brick	1	1.1	-	17	-	0.63	-
LECA block	4	1.3	0.77-1.6	38	23-47	1.3	0.90-1.8
SL20 silica white brick	5	3.1	2.7-3.8	48	41-58	1.8	1.6-2.2
F54 flint and lime buff brick	5	2.2	1.4-2.7	33	22-43	1.1	0.75-1.2
F50 heather silica brick	5	2.6	2.2-3.1	40	33-48	1.4	1.1-1.6
Silica brick of unknown manufacture	12	9.2	7.7-11	130	120-150	5.5	4.8-6.3
Appleby rural red rustic brick	5	1.8	1.5-2.4	30	25-40	1.0	0.87-1.4

Table C1 (cont.)

Sample	No.	Radon-222 emanation rates					
		$\text{Bq m}^3 \text{s}^{-1} \times 10^{-3}$		$\text{Bq m}^{-2} \text{s}^{-1} \times 10^{-6}$		$\text{Bq kg}^{-1} \text{s}^{-1} \times 10^{-6}$	
		mean	range	mean	range	mean	range
Beare Green pastone clay brick	5	0.90	0.68-1.1	13	10-17	0.53	0.43-0.58
Beare Green Weald hand made light multicl原因ay brick	5	1.1	0.83-1.6	18	13-25	0.63	0.48-0.98
Blaby golden blende hand made clay brick	5	0.95	0.68-1.10	10	7.3-12	0.53	0.38-0.61
Dorking multicolour stock clay brick	3	0.32	0.16-0.50	3.6	2.0-5.7	0.15	0.08-0.24
Ibstock beacon Sahara multicl原因ay brick	5	0.65	0.52-0.85	7.8	1.4-12	0.42	0.32-0.54
Ibstock red rustic multicl原因ay brick	5	1.2	1.0 -1.3	15	13-17	0.63	0.56-0.70
Northcot multicl原因ay brick	5	0.70	0.55-0.86	12	9.2-14	0.38	0.30-0.47
LBC dapple light clay brick	5	0.95	0.73-1.1	14	12-16	0.63	0.54-0.71
LBC Cotswold clay brick	4	0.95	0.61-1.2	11	6.6-14	0.56	0.35-0.73
Blaby Winsdor grey brick	5	0.88	0.55-1.2	13	8.6-17	0.53	0.34-0.66
Chailey stock clay brick	5	1.3	1.1 -1.8	15	12-23	0.81	0.64-1.1
Elliot light red brick	5	1.2	0.66-1.5	12	8.5-14	0.47	0.33-0.60
Kirton brown clay brick	5	0.97	0.58-1.4	10	6.1-16	0.59	0.39-0.80
Kirton minster grey hand made brick	5	1.2	0.88-1.4	12	8.9-15	0.64	0.48-0.78
Otterham second hard stock clay brick	5	1.4	1.2 -1.6	21	18-24	0.92	0.82-0.99
Severn Valley orange multicl原因ay brick	5	1.1	0.87-1.2	13	11-15	0.66	0.53-0.73
Armitage class B brick	5	1.2	0.97-1.4	13	10-15	0.48	0.39-0.58
Baggeridge best blue brick	5	0.89	0.68-1.0	15	12-17	0.40	0.31-0.44
Baggeridge red rustic brick	5	1.4	1.0-1.6	21	14-22	0.81	0.60-0.92
Ludlow crushed granite brick	4	7.8	5.2-11	130	88-180	3.6	2.4-5.2
9" Fyfe-stone block (grey)	3	22	19-27	630	540-790	9.7	7.9-12
9" Fyfe-stone block (pink)	3	20	19-21	540	510-580	9.0	8.8-9.5

Table C2  
Manufacturing process and constituents of a selection  
of materials given in Table C1

Material	Details
Blaby Windsor grey brick	<p><u>Main constituent:</u> Keuper Marl (late Triassic in age). No information supplied as to analysis of marl used.</p> <p><u>Process:</u> fired to a temperature of 1060°C using natural gas.</p> <p><u>Water absorption:</u> 27% by weight.</p>
Blaby golden brown brick	As for Blaby Windsor grey, but with different facing sand.
Beare green wealde hand made light multicl原因 brick	<p><u>Main constituent:</u> Weald clay having approximate composition, 67% SiO<sub>2</sub>; 17% Al<sub>2</sub>O<sub>3</sub>; 4.8% Fe<sub>2</sub>O<sub>3</sub>; 0.71% CaO; 0.62% MgO; 1.7% K<sub>2</sub>O and 0.27% Na<sub>2</sub>O, with a 6.3% ignition loss.</p> <p><u>Additive:</u> solid fuel</p> <p><u>Process:</u> soft mud</p> <p><u>Water absorption:</u> 16% by weight</p>
Beare Green pastone	As for Beare Green Wealde hand made light multicl原因, but with the addition of a small quantity of TiO <sub>2</sub> to the facing sand.
Ibstock beacon Sahara multicl原因 brick	<p><u>Main constituent:</u> Keuper Marl as described for the Appleby rural red rustic, but using buff clay having the following composition, 55% SiO<sub>2</sub>; 0.6% TiO<sub>2</sub>; 12.1% Al<sub>2</sub>O<sub>3</sub>; 4.7% Fe<sub>2</sub>O<sub>3</sub>; 6.3% CaO; 4.9% MgO; 5.0% K<sub>2</sub>O and 0.3% Na<sub>2</sub>O, with 11.4% ignition loss.</p> <p><u>Additives:</u> pond slurry analysed as 30% moisture, 21% ash, 1.5% sulphur, 0.9% chlorine. Barium sulphate is added as a slurry at 0.1% by weight.</p> <p><u>Process:</u> wire cut.</p> <p><u>Water absorption:</u> 21% by weight.</p>
Ibstock red rustic multicl原因 brick	As for the Appleby rural red brick, but with the omission of the coke breeze.
Appleby rural red rustic brick	<p><u>Main constituent:</u> Keuper Marl laid down with near horizontal seams of red and grey/green mudstone combined with siltstone bands and sandstone. The red clay is used for this brick and has the following composition. 60.7% SiO<sub>2</sub>; 0.7% TiO<sub>2</sub>; 12.5% Al<sub>2</sub>O<sub>3</sub>; 5.2% Fe<sub>2</sub>O<sub>3</sub>; 2.3% CaO; 2.8% MgO; 4.8% K<sub>2</sub>O and 0.3% Na<sub>2</sub>O.</p> <p><u>Additives:</u> coke breeze (as fuel) analysed as 15% moisture, 10% ash, 1.25% sulphur, 73% fixed carbon and 2% volatiles. Barium carbonate added as 0.1% by weight to prevent 'scumming' and effluorescence.</p> <p><u>Process:</u> wire cut</p> <p><u>Water absorption:</u> 16% by weight</p>
Northcot multicl原因 brick	<p><u>Main constituent:</u> clay of the Lower Lias from the Wellaoe Quarry, Blockley, Gloucestershire.</p> <p><u>Additive:</u> none given.</p>

Table C2 (Cont.)

Material	Details
Chailey stock clay brick	<p><u>Main constituent:</u> Weald clay having approximate composition, 75% <math>\text{SiO}_2</math>; 9.8% <math>\text{Al}_2\text{O}_3</math>; 4.3% <math>\text{Fe}_2\text{O}_3</math>; 1.5% <math>\text{CaO}</math>; 0.31% <math>\text{MgO}</math>; 0.47% <math>\text{K}_2\text{O}</math> and 2.2% <math>\text{Na}_2\text{O}</math>, with a 5% ignition loss.</p> <p><u>Additive:</u> solid fuel.</p> <p><u>Process:</u> soft mud.</p> <p><u>Water absorption:</u> 9% by weight.</p>
Kirton brown clay brick	<p><u>Main constituent:</u> Kauper Marl fired to temperature of <math>1050^\circ\text{C}</math> using liquid petroleum gas.</p> <p><u>Additive:</u> none stated.</p> <p><u>Process:</u> extruded wire cut.</p> <p><u>Water absorption:</u> 25% by weight.</p>
Kirton Minster grey brick	<p>As for Kirton brown clay brick, but with different facing sand.</p>
Otterham second hard stock	<p><u>Main constituent:</u> Kent brick earth containing approximately 69% <math>\text{SiO}_2</math>; 11% <math>\text{Al}_2\text{O}_3</math>; 4% <math>\text{Fe}_2\text{O}_3</math>; 5% <math>\text{CaO}</math>; 2% <math>\text{MgO}</math>; 2% <math>\text{K}_2\text{O}</math> and 1% <math>\text{Na}_2\text{O}</math>, with an ignition loss of 6.5%.</p> <p><u>Additives:</u> solid fuel, chalk. The chalk is added purely to give a yellow colour to the finished product.</p> <p><u>Process:</u> soft mud.</p> <p><u>Water absorption:</u> (of the fired brick): 23% by weight.</p>
Baggeridge best blue, grade A	<p><u>Main constituent:</u> Etruria Marl quarried from a bed just south of the northern outcrop of the north Warwickshire coal beds.</p> <p><u>Additives:</u> none except water.</p> <p><u>Process:</u> extrusion. Final stage of firing carried out in a reducing atmosphere to develop blue colour.</p> <p><u>Water absorption:</u> approximately 3.3% by weight.</p>
SL20 brick	<p><u>Main constituents:</u> Ryarsh quartz greensand from Ryarsh, Kent. 11.6% of hydrated lime (from Blue Circle Industries) and 1-2% of mains water are added. These being percentages by weight of total silicious aggregate.</p> <p><u>Process:</u> not stated.</p> <p><u>Water absorption:</u> not stated.</p>
F54 buff brick	<p><u>Main constituents:</u> 53% durite (a calcined flint), 27% sea-dredged quartz and 20% Ryarsh quartz greensand.</p> <p><u>Additives:</u> 8.8% hydrated lime, 0.5% yellow synthetic iron oxide and 1-2% of mains water. These percentages are by weight of total weight of the silicious aggregate.</p> <p><u>Water absorption:</u> not stated</p>
F50 heath brick	<p>The same as the F54 buff brick with the exception that there is no yellow iron oxide pigment added.</p>

As can be seen from Table C2 the F54 and F50 bricks are of similar composition, as are the two Beare Green bricks, the Blaby bricks and the Kirton bricks. Although no details are available on the constituents of the LBC dapple light and the LBC Cotswold bricks these are the same fletton type brick with different facing sands. Inspection of Table C1 indicates that these pairs of bricks do have similar radon-222 emanation rates.

The emanation of radon-222 from materials is affected by such parameters as humidity and atmospheric pressure. To investigate the repeatability of the emanation rate measurements a limited number of specimens were measured again some two months after the first measurement. The results for the three 4-inch concrete blocks are given in Table C3.

Table C3  
Repeated measurements of radon-222 emanation rates  
of 4 inch concrete blocks

Block	Radon-222 emanation rates					
	1st measurement			2nd measurement		
	$\text{Bq m}^{-3} \text{ s}^{-1}$ $\times 10^{-3}$	$\text{Bq m}^{-2} \text{ s}^{-1}$ $\times 10^{-6}$	$\text{Bq kg}^{-1} \text{ s}^{-1}$ $\times 10^{-6}$	$\text{Bq m}^{-3} \text{ s}^{-1}$ $\times 10^{-3}$	$\text{Bq m}^{-2} \text{ s}^{-1}$ $\times 10^{-6}$	$\text{Bq kg}^{-1} \text{ s}^{-1}$ $\times 10^{-6}$
1	6.8	200	3.5	6.5	200	3.4
2	7.5	220	3.8	6.7	200	3.4
3	7.6	220	3.8	6.7	200	3.4

Table C3 suggests that individual measurements of radon-222 emanation rates produce results that are within 10% of the average value. The reason for the remarkably close agreement between the specimens at the second measurement stage is unexplained.





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