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INDOOR RADON CONCENTRATIONS AND BUILDING MATERIALS CONTROL OF AIRBORNE RADIOACTIVITY

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ABSTRACT

The contribution to the airborne radioactivity from the building materials can be predicted by measurements of exhalation rates of samples placed in closed containers. The effect of sample dimensions, porosity and dead space on the back diffusion is discussed, and it is shown that the back diffusion will usually not change the measured exhalation rates with more than 10-15%.

Efforts on reducing ventilation rates in order to save energy can lead to unacceptable high levels of radon and radon daughters in the indoor environment.

The major radon source for indoor air is often the soil, from which radon can diffuse through the base of the house. The possibility of reducing the diffusion rate by proper composition of the floor and base material is mentioned.

Finally the results of an investigation on reducing the radon daughter concentrations by circulation and filtration are reported. It is shown that a passage of the air through an ordinary filter with a rate of 1-2 h^{-1} may reduce the radon daughter level with a factor of 5-10.

KEYWORDS

Radon; building materials and radon; radon daughters; control of airborne radioactivity.

INTRODUCTION

About sixty years ago it was established that ionizing radiation originating from uranium ore was a major cause of the high incidence of lung cancer among workers in underground mines. In 1924 it was shown that the radiation was associated with the inert gas radon, which is the first daughter product of radium, but it lasted almost another 30 years before it was demonstrated (Harley, 1952), that the airborne short-lived daughter products of radon rather than the mother product were the real source of the major part of the radiation dose.

For several years this kind of radiational exposure was supposed to be an exclusively occupational hazard. By the end of the fifties, however, it was realized, that elevated levels of airborne radioactivity might also occur in ordinary overground houses.

The sources of radon in indoor air are the building materials and the soil under the houses.

Many building materials as well as the soil will contain trace amounts of radium, often in the order of pg/g. The radium itself is bound to the matrix of the material and the radiation from the radium can usually be ignored from a health hazards point of view.

When radium decays, however, the atoms of the daughter product, radon, which as indicated is an inert gas, may diffuse out of the material and mix with the sorrounding air. If the radon atoms are inhaled, they may decay in the respiratory tract with damage of the epithel as a result. Although radon is somewhat soluble in water, and a certain enrichment therefore may take place in the lungs, the major parts of the radon atoms are exhaled again before they decay.

The daughter products of radon, RaA, RaB, RaC and RaC', are chemically very active materials which attach readily to condensation nuclei in the air, and, when inhaled, are very likely to deposit somewhere in the respiratory tract.

The concentration of radon as well as of its airborne daughter products are measured in Bq/m^3 or traditionally in pCi/ℓ. The radiation hazard is connected with the alpha radiation, and since two of the daughter products, RaA(^{218}Po) and RaC'(^{214}Po), are alpha emitters, the same radiation burden can arise from many combinations of concentrations of the individual daughters. In order to take this fact into account a special unit, the working level, WL, is defined as

$$WL = 0.00103\ RaA + 0.00507\ RaB + 0.00373\ RaC \qquad (1)$$

where RaA, RaB and RaC are the concentrations in pCi/ℓ.

The definition also involves that 100 pCi/ℓ of radon in equilibrium with its daughters correspond to 1 WL.

The working level thus gives in one figure a measure of the total radiational burden of the respiratory tract.

There are no generally accepted maximum permissible levels of airborne radioactivity outside the occupational situation, but it is usually agreed that levels up to 3 pCi/ℓ of radon in radioactive equilibrium with its daughter products can be accepted for rooms where people spend up to 40 hours per week and levels up to 1 pCi/ℓ for living rooms, where people may spend up to 160 hours per week.

The limits should, however, be related to the daughter products, rather than to the radon itself, and should therefore be 0.03 WL and 0.01 WL respectively, and since radon only rarely is in equilibrium with its daughter products, this may correspond to radon concentrations of 6 and 2 pCi/ℓ.

EXHALATION OF RADON

The diffusion, or exhalation, of radon from a given material can at constant pressure be considered as a material parameter. The exhalation properties can be studied by enclosing samples of the materials in closed containers and follow the growth of radon activity in the container by analysis of air samples.

If the radon concentration in the container (atoms per unit volume) at equilibrium is A_o, the dead space volume of the container V_d, the radon exkaling surface S, and the decay constant of radon λ, then the net exhalation rate E' at equilibrium can be written (Jonassen and McLaughlin, 1978)

$$E' = \frac{\lambda A_o V_d}{S} \qquad (2)$$

This figure, however, will be lower than the free exhalation rate E corresponding to exhalation into an empty space, or into an ordinary room.

The difference is caused by the so-called back diffusion, which will slow down the exhalation rate from the material as the activity (concentration) in the container grows.

The magnitude of the difference depends upon a) the dimensions of the sample relative to the diffusion length of the material, and b) the ratio between the dead volume of the container and the void volume of the sample, i.e. the product of the porosity and the sample volume. If the dead space is chosen as approximately twice the sample volume, then for most building materials with porosities below 0.5 the exhalation value determined from eq. (2) will not differ from the free exhalation value by more than 10-15% at the most.

In the first column of Table 1 are shown the results of measurements of exhalation rates from a series of commonly used building materials

TABLE 1 Exhalation rates and contributions to radon concentrations for various building materials

Material	Exhalation rate		Radon concentration	
	$\frac{\text{atoms}}{m^2 s}$	$\frac{\text{pCi}}{m^2 s}$	R_0 $\frac{\text{pCi}}{\ell}$	$R_{0.5}$
chipboard fiberboard gypsum board (nat. gypsum)	<1	$<5 \cdot 10^{-5}$	$<5 \cdot 10^{-2}$	$<8 \cdot 10^{-4}$
rockwool bricks	2	$1.1 \cdot 10^{-4}$	$1 \cdot 10^{-1}$	$1.6 \cdot 10^{-3}$
light weight concretes (dan. origin)	20-30	$1.1\text{-}1.7 \cdot 10^{-3}$	1-1.5	$1.6\text{-}2.4 \cdot 10^{-2}$
ordinary concretes (danish dep.)	130-180	$7\text{-}10 \cdot 10^{-3}$	7-10	$1.1\text{-}1.5 \cdot 10^{-1}$
gypsum tiles (chemogyps.)	800	$4.5 \cdot 10^{-2}$	43	$6.4 \cdot 10^{-1}$
alum shale light weight concrete	1400-4000	$8\text{-}22 \cdot 10^{-2}$	76-200	1.1-3.2

If a material with the exhalation rate E is being used in a room the contribution to the radon concentration from the material is given by

$$R_n = \frac{\lambda}{\lambda+n} \cdot \frac{S}{V} \cdot E \qquad (3)$$

where S is the exhaling area, V the volume of the room, n the ventilation rate and λ the decay constant of radon.

If all surfaces of a room are covered with the exhaling material, then $\frac{S}{V} \sim 2\ m^{-1}$.

The corresponding radon concentrations R_o for an unventilated room and $R_{0.5}$ for a room with a ventilation rate of $0.5\ h^{-1}$ are shown in the second column in the Table.

These concentrations are the maximum contributions from the materials under the given ventilation conditions.

The figures in Table 1 are related to the surface area of the samples, and it is assumed that the values obtained from the samples can be scaled up to the dimensions of a room. In cases where the materials for instance are being used as boards or tiles, or more generally where the dimension perpendicular to the exhaling surfaces is small compared to the diffusion length, this procedure may underestimate the actual exhalation rate, and it would be more appropriate to express the exhalation rate relative to a unit mass of the material.

The fact that usually not all surfaces of a room are covered with the (same) radon exhaling material may reduce the concentrations i column 2, Table 1 somewhat. Because of influx of radon by diffusion from the soil through the base of the house the total concentrations of radon in a house built of a given material may, on the other hand, be considerably higher than appears from Table 1.

This is in correspondence with the findings from many countries (Cliff, 1978, Swedjemark, 1977) where radon concentrations in tens of picocurie per liters have frequently been reported in ordinary houses. The concentrations of the daughter products are always lower, but may though often attain unacceptably high levels.

CONTROL OF AIRBORNE RADIOACTIVITY

The simplest and most effective way of lowering the level of airborne radioactivity in a room is by increasing the ventilation rate. This follows from eq. (3) which can be rewritten

$$R_n = \frac{\lambda}{\lambda+n} \cdot R_o \qquad (4)$$

where R_o is the radon concentration corresponding to an unventilated room, including the contribution from the walls as well as that from the soil underneath the house.

Since $\lambda = 7.5 \cdot 10^{-3} h^{-1}$ a ventilation rate of 0.5 h^{-1} will lower the radon concentration to 1.5% of the unventilated value.

An increase in the ventilation rate in order to lower the radon level will, however, be in variance with attempts to save energy by improving the insulation of the house. It is therefore of considerable interest to investigate other means of reducing the level of radon and radon daughters in the indoor air.

It is in principle possible to reduce the exhalation of radon from the surfaces of a room by a treatment with a pore filling agent. Since the larger part of the radon flux often comes from or through the floor most of the effort has been directed towards developing suitable floor sealants. It appears that sealants of the epoxy type, when properly applied, can reduce the radon concentration in the room to almost background levels (Culot, Olson and Schiager, 1978). An adverse effect is that radon may be trapped back of the sealant layer and hereby increase the level of γ-radiation in the room. It should be stressed that cracks and joints between walls and the floor may often act as major radon feed lines and should therefore be given special attention in the sealing process.

An alternative to stopping the radon by an external sealant is to use a floor material which offers a high resistance to radon diffusion. Some floor materials like many types of concrete are rather permeable to radon and will therefore only offer little resistance to the diffusion. It is therefore of interest to find if it is possible to change the diffusion rate through the material for instance by changes in the hardening process. Attempts in this direction are presently being made.

Even if it is difficult to control the feed of radon to the air of a given room it may be possible to treat the air in such a way that radon and/or its airborne daughter products are removed from the air totally or in part.

Radon itself is known to be adsorbed by the surface of materials like active charcoal, but the adsorption efficiency is rather low at room temperature.
In order to produce a significant drop in the radon level by passing the air through uncooled charcoal filters, it is therefore necessary to use very bulky and impractical filter arrangements. It is, however, possible that even a moderate lowering of the filter temperature may increase the adsorption efficiency to practical levels. This possibility has, to the knowledge of the author, not yet been investigated.
The daughter products of radon on the other hand have a high probability of being retained by filters or adhering to surfaces by contact. In order to investigate the possibilities of removing the radon daughters by a mechanical treatment of the air a series of experiments were performed at the Technical University of Denmark. The room used for the measurements is a basement room with walls, floor and ceiling made of concrete. The average exhalation rate of radon is 380 atoms/m^2·s, the volume of the room is 324 m^3 and the surface area is 360 m^2 (McLaughlin and Jonassen, 1978). The natural air exchange rate of the room is about 0.2 per day (corresponding to radon free air). The long term average radon activity is about 5-10 pCi/ℓ. Simultaneous measurements of the activity of radon and its daughter products were done in the period Sep.-Nov. 1979. Atmospheric pressure, temperature, relative humidity and condensation nucleus concentration were also determined (Jonassen, 1980). Measurements were taken in different states or during different treatments of the air: 1) undisturbed or static air, 2) air passing through a fan system with no direct filtration at a flow rate of 1200 m^3/h (or an exchange rate for the room of $\sim 10\cdot10^{-4}s^{-1}$) and 3) air filtered at 400 m^3/h ($\sim 3\cdot10^{-4}s^{-1}$) and 1200 m^3/h ($\sim 10\cdot10^{-4}s^{-1}$).

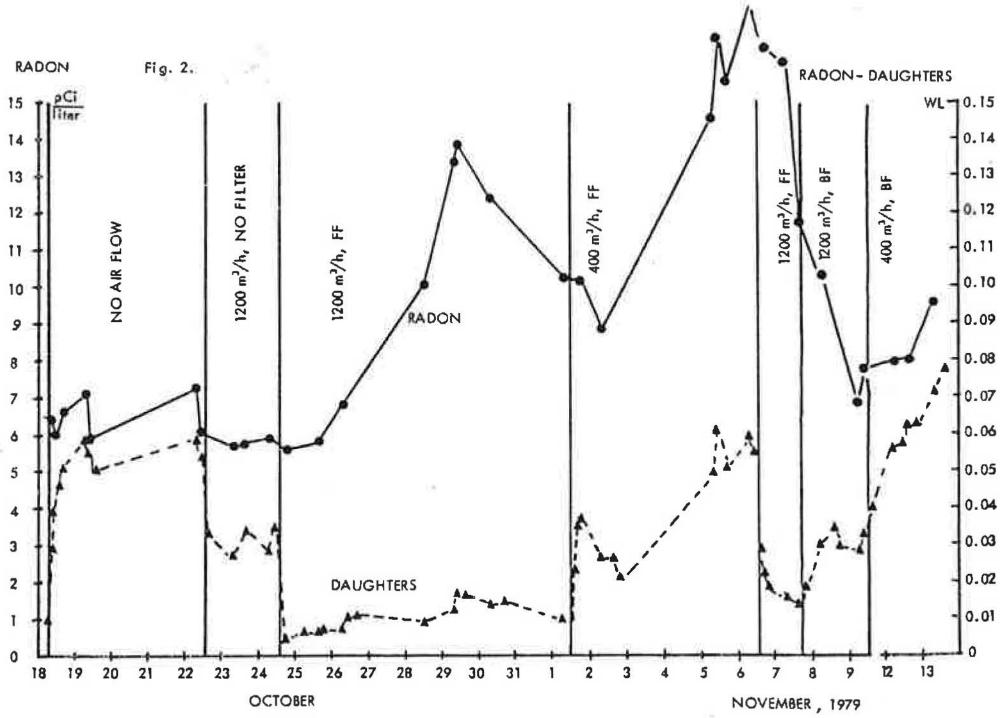

Fig. 1. Radon concentration and working level for various treatment of the air.

Some of the results are shown in Fig. 1. The solid line shows the radon concentration (left hand scale) and the dotted line the working level of the radon daughters (right hand scale). The filters used were a so-called basic filter (BF) and a fine filter (FF) of the type Comfit-85, and only the results obtained with the latter will be dealt with in the discussion of the effects of filtration.
It appears from Fig. 1, that the radon activity is not being lowered by any of the treatments. The increase in radon concentration in the period Oct. 25-29 from 5.5 to 14 pCi/ℓ accompanied a drop in atmospheric pressure from 769 to 758 mmHg. In the period Nov. 2-6 the corresponding figures were 9 to 19 pCi/ℓ and 760 to 733 mmHg. This is in good agreement with earlier results (Jonassen, 1975).
In order to characterize the effect on the daughter products of the different treatments of the air the so-called equilibrium factor F defined as

$$F = 100 \cdot \frac{WL}{Rn} \tag{5}$$

is used, where Rn is the radon concentration in pCi/ℓ.
The results are shown in Table 2. The figures in brackets are the standard deviation of a single determination of the figure directly above

TABLE 2 Effect of circulation and ventilation

Treatment of the air	Air exchange rate $10^{-4}s^{-1}$	Equil. factor F	Condensation nuclei cm^{-3}
static	0.02	0.84 (0.08)	15 000
circul. 1200 m^3/h	10	0.53 (0.05)	10 000
filtrat. 400 m^3/h	3	0.32 (0.04)	600
filtrat. 1200 m^3/h	10	0.12 (0.02)	100

It appears that even in the undisturbed state of the air the radon daughters are not in total equilibrium with the parent product, radon. This is due to plate out to the walls mainly caused by diffusion and random movement. A closer analysis (Jonassen, 1980) shows that only RaA and, to a smaller extent, RaB are affected by the plate out.
When the air is circulated through the fan system with an air exchange rate of $10 \cdot 10^{-4}s^{-1}$ the equilibrium factor drops to 0.53. Measurements of the daughter concentrations in front and back of the fan system, however, show no change in the concentrations by the passage of the fan, and the effect is therefore believed to be caused by an increased plate out (of RaA). This increase may be caused by the stirring of the air, making more RaA-particles come close to the walls, but can also be caused by the decrease in the nucleus concentration resulting in a larger fraction of RaA-atoms being unattached and thus more prone to diffusion to the walls.
When the air is passed through filters the working level decreases with increasing filtration rate. An analysis of the relative concentrations of the individual daughters (Jonassen, 1980) reveals that RaB and RaC are removed with a rate which is

rather close to the filtration rate, while the removal rate of RaA is many times higher. This is undoubtedly due to an increase in the unattached fraction of RaA caused by the large decrease in the nucleus concentration. The results reported here are to be considered as preliminary. On one hand it appears that a circulation or filtration with a moderate rate may cause a significant decrease in the working level in a room. On the other hand a filtration will increase the unatteched fraction of the radon daughters, and thus changing the radiational impact of the inhaled air on the respiratory tract.

In the investigation reported above no attempts were made to measure the unattached fraction of the radon daughters. Such measurements are, however, necessary in order to give a complete characterization of the radiological properties of the air and will be part of the future work on control of airborne radioactivity.

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