THE IONIZING RADIATION IN DWELLINGS RELATED TO THE BUILDING MATERIALS

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Abstract

When proposing norms for the radioactivity in dwellings the aim is to limit the absorbed dose in humans. For practical reasons, however, it is necessary to use limits for the concentration of the radionuclides in the building materials. At the discussions of standards for radioactivity in dwellings the formula

$$m = \frac{C_{Th}}{260} + \frac{C_{Ra}}{370} + \frac{C_K}{4.8 \times 10^3}$$

has been used. C is expressed in Bq/kg[a)] of thorium-232, radium-226 and potassium-40. Krisiuk et al. have shown that if m in the formula has the value 1.0 the absorbed dose in the gonads would not exceed 1.5 mGy/y[b)].

Experimentally determined values were used to investigate the absorbed dose or concentration of radon and daughter products after normalizing the activity concentration in the building materials to m = 1. The exposure rates, the concentrations of radon and daughter products and the ventilation rates have been determined in seven groups of houses built in the early 1970s in central Sweden. The activity concentrations in the building materials used at the time for building the houses were measured earlier.

The absorbed dose in the gonads normalized to m = 1 was, as expected, found to be lower than the theoretical value, 1.5 mGy/y, according to the model. For the multi-family houses the values were about 1.0 mGy/y and for the single-family houses about half that value.

The concentrations of radon in equilibrium with the daughter products normalized to one air change per hour and to the maximum concentration of radium-226 in the building material according to the model were found to be in accordance with the theoretical value, about 100 Bq/m^3, for the multi-family houses. No significant difference from this value was found for the single-family houses despite the fact that the amounts of stony building materials were less than in the multi-family houses.

a) 1 Bq/kg ≈ 0.027 pCi/g
b) 1 mGy/y = 100 mrad/y

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Introduction

When proposing norms for the radioactivity in dwellings the aim is to limit the absorbed dose in humans. For practical reasons, however, it is necessary to use limits for the concentration of the radionuclides in the building materials. The correlation between the absorbed dose in humans and the concentration of the radionuclides in the building materials is very complicated. Some experimental correlations regarding levels of gamma radiation and radon are examined below.

The evaluation of the external doses is treated under one heading and the concentrations of radon and radon daughters under a separate heading.

Measurements

In seven groups of houses in the town Gävle in central Sweden the concentrations of radon and daughter products have been determined in conjunction with the determination of the air changes during winter and early spring (Ref 1). The exposure rates were also determined. The investigation comprised the conditions of 1975-1976[a] and for the multi-family houses also the conditions after adjusting the air intake to the norms of the Swedish Building Code of 1975.

Three groups of multi-family houses and four groups of single-family houses built of various kinds of building materials and with various ventilation systems were investigated in the winter and early spring of 1976. Table 1 shows the building materials of the houses, the ventilation systems and also summarizes the measurement results. All the groups of the multi-family houses and two of the groups of the single-family houses had mechanical exhaust ventilation. The other two groups had natural ventilation. The single-family houses without a cellar were built on a slab of ordinary concrete.

The occupants were asked to keep the windows shut from the evening prior to the measurements, but of course the doors had to be opened and the frequency of the door opening differed from family to family.

Air samples were taken with evacuated containers and measured the same day in the ionizing chambers which have been used for the determination of the radon concentration in non-uranium mines in Sweden for several years (Ref 3). Filter samples have been taken for determining the shortlived radon daughters, also with the same method as used for the mines. The total alpha activity of the filters was measured with a zinc sulphide detector. The activity was calculated using Kusnetz's method (Ref 4). The minimum detectable radon concentration ($3\ \sigma_{background}$) was about 26 Bq/m^3 [b]. For the radon daughters the figure was about 2 Bq/m^3.

The measurements of the air changes have been made with a tracer gas method. N_2O is let out in the dwelling and an analyzer records the decrease of the concentration of the gas.

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a) Earlier, Swedish dwellings built before 1946 have been investigated with regard to the gamma radiation and the concentrations of radon-222 and radon-220 (Ref 2).

b) 1 $Bq/m^3 \approx 0.027$ pCi/l.

The activity concentrations of the building materials of the kind used at the time of building the houses have been summarized in Table 2 (Ref 5, 6, 7). The activity concentration average for the concrete has been calculated from measured concentrations in samples of ballast material and cement. The ballast material was taken from gravel-pits in the Gävle area, the cement samples were an average from all the manufacturers in Sweden. For the remaining materials, averages have been obtained using samples from factories situated in Sweden.

The absorbed dose in humans from the external radiation related to the concentration of the radionuclides in the building material

Krisiuk et al. (Ref 8) after calculations have concluded that an absorbed dose in the gonads of 1.5 mGy/y [a] (150 mrad/y) from external gamma radiation corresponds to a concentration of 260 Bq [b] thorium-232 per kg or 370 Bq radium-226 per kg or 4.8 x 10^3 Bq potassium-40 per kg of the building material.

These concentration values are roughly related to the inverse of the exposure rate constants for the nuclides. Usually the three nuclides are present at the same time in the building material and according to the above source, the absorbed dose in the gonads will not exceed 1.5 mGy/y if

$$\frac{C_{Th}}{260} + \frac{C_{Ra}}{370} + \frac{C_K}{4.8\times10^3} = 1 \qquad (1)$$

where C is expressed in Bq/kg of thorium-232, radium-226 and potassium-40. The model takes no consideration to windows or doors or to the fact that walls are not infinitely thick. The absorbed dose from the external gamma radiation is therefore lower in reality.

An estimate of how much lower the radiation dose can be has been made in the following way. The expression to the left of the equality sign in equation 1 has been called m

$$m = \frac{C_{Th}}{260} + \frac{C_{Ra}}{370} + \frac{C_K}{4.8\times10^3} \qquad (2)$$

m is then approximately proportional to the gamma activity in the building material. From the measured activity concentrations in various building materials m has been determined as an average for each group of houses with the exception of one group. The radiation doses measured in the dwellings have then been normalized to m = 1.

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a) 1 mGy/year = 100 mrad/y

b) 1 Bq/kg ≈ 0.027 pCi/g

Multi-family houses

In the groups of houses investigated, equation (1) is applicable for the multi-family houses. Almost all of the building materials used in these houses contain radionuclides which contribute significantly to the radiation dose. For the multi-family houses it is therefore appropriate to weight the content of building materials for the various activity concentrations to get an estimate of m. The material of a house built mainly of concrete screens out most of the outdoor radiation. Therefore, the measured radiation dose comes from the building materials. After the calculation of the dose rate for $m = 1$ and subtracting the average outdoor dose rate, the absorbed dose in the gonads was calculated to 0.95 ± 0.24 mGy/y from the building materials of the multi-family houses (Table 3a).

0.24 mGy/y is an estimate of the extreme limit in the absolute determination of the normalized value. It is composed by two different types of errors, the standard deviation for the dose rate and the error in m. The later error can be seen as systematic, is an extreme limit and is dominating. The fact that it is large, depends on the many approximations necessary partly because it has not been possible to investigate samples of building materials directly taken from the buildings, partly according to the difficulties associated with the weighting of the various building materials in the same house. The building materials are similar in the multi-family houses and therefore the error in m is of most importance on the determination of the absolute normalized average. The range between the averages for the groups of multi-family houses is only 0.06 mGy/y which is in accordance with the error in the average about 0.01 mGy/y, when the error in m is not taken into consideration.

Single-family houses

For single-family houses the major part of the building material more seldom contains activities of importance to the radiation dose. The only rooms in this investigation where equation (1) is valid, are the cellars in group No. 5, the ground floors in group No. 6 and No. 7.

The walls in the single-family houses are of lighter material than the multi-family houses and therefore screen the outdoor radiation to about half (Ref 9). Thus when normalizing the measured dose rate to $m = 1$, half of the outdoor dose rate has been subtracted before and the other half after the normalizing.

The cellars of group No. 5 were surrounded by stone material; heavy concrete in the floor and the ceiling and aerated concrete based on alum shale in both the outer and the inner walls. The absorbed dose rate, 0.62 ± 0.12 mGy/y, for $m = 1$ does not differ significantly from the values for the multi-family houses.

The ground floor in group No. 6 was also surrounded by stone material, heavy concrete in the floor, aerated concrete based on sand in the ceiling and aerated concrete based on alum shale in both the outer and the inner walls. For $m = 1$ the absorbed dose rate of 0.53 ± 0.17 mGy/y was found. It differs significantly from the multi-family houses but not from the cellars in group No. 5. The lower value compared to the multi-family houses is probably due to the lower density of the aerated concrete ($0.5 \cdot 10^3$ kg/m^3) compared to heavy concrete ($2.3 \cdot 10^3$ kg/m^3).

The materials in the houses in group No. 7 were heavy concrete in the floor and the facade brick of clay together with a wood construction and insulating material in the outer walls. The ceiling was of wood and insulating stone wool, which does not give any contribution of importance to the radiation. For m = 1 the absorbed dose rate was found to be 0.42 $\pm$ 0.10 mGy/y. It is possible that this value has been somewhat underestimated because the activity concentration in clay brick (Table 2) is not representative for Sweden and could be overestimated. The dose rate normalised to m = 1 is significantly lower than the corresponding value for the multi-family houses.

The concentration of radon and radon daughters related to the concentration of radium in the building material

If the only radionuclide contained in the building material is radium-226, equation (1) means that 370 Bq ^{226}Ra/kg corresponds m = 1. Krisiuk et al. showed that 370 Bq ^{226}Ra/kg material gives an increase of the concentration of radon not exceeding 110 Bq/m^3 air in equilibrium with the daughter products when the ventilation rate is about one air change per hour.

The concentration of radium has been weighted for the weights of the various building materials in the dwelling. The concentration of radon and radon daughters calculated for one air change per hour has then been related to the weighted concentration of radium in the building material.

Multi-family houses, group No. 1 - 3

If the concentration of radium-226 in the building material was normalized to 370 Bq/kg and the air change to 1 change per hour, the concentrations of radon and daughter products in the air were found to be 270 $\pm$ 90 Bq/m^3 and 81 $\pm$ 33 Bq/m^3 respectively for the multi-family houses. This is in accordance with the calculations made by Krisiuk et al.

The ground floor dwellings have been excluded in the above averages, because the concentrations of radon and daughter products were significantly higher than in the remaining dwellings in the investigated groups of houses. For group No. 1 the higher concentrations could be explained by the presence of aerated concrete based on alum shale in the ground floor dwelling. For group No. 3, however, no such explanation was found and the higher radon levels could have been caused by several factors. A higher radon exhalation from a basement slab than from a wall made of the same material is quite possible. A part of the radon from the underlying ground passes through the basement slab (Ref 10). Light weight aggregate used in an edge beam contains higher concentrations of radium-226 than the concrete from the town Gävle and perhaps contribute to the radon level in air. The walls in the ground floor may be thicker than in the dwellings higher up in the house.

Tap water can be one source for the radon in the air of a building. All of the investigated groups of houses used the same source for the tap water. The radon concentration was found to be 12000 $\pm$ 2000 Bq/m^3 water (1 σ). The highest concentration of radon which could be expected in the air from this concentration of radon in the water is about 7 Bq/m^3 air (Ref 11).

Single-family houses without aerated concrete based on alum shale, groups Nos. 4 and 7

For all houses, where the material mainly responsible for the radon concentration in the house is the concrete, the same value for the weighted concentration of radium-226 has been used. However, the contents of concrete vary between the various groups of houses. The dwellings in the multi-family houses are almost entirely surrounded by concrete, while the investigated single-family houses have concrete only in the basement slab or in the cellar. Despite that fact, the concentration of radon daughters, calculated for one air change per hour, in the houses of group No. 4 is insignificantly lower than in the multi-family houses.

The houses in group No. 7 do not have any cellars. Beside the concrete in the basement slab, the facade brick wall exhales radon. The wall of facade brick is small but the concentration of radium-226 can be higher than in the concrete from Gävle. Therefore, the facade brick can be of importance as a radon source. The concentration of radon and radon daughters for one air change per hour and for 370 Bq ^{226}Ra per kg of the building material is insignificantly higher than for the multi-family houses.

That the concentrations of radon and daughter products normalized to m = 1 are of the same order of magnitude in the single-family houses as in the multi-family houses despite the lower amount of stone material is probably due to the fact that the single-family houses are in close contact with the ground. The concentrations of radon and daughter products were about twice as high in a ground floor dwelling as in the remaining dwellings higher up in a group of multi-family houses (group No. 3), where the building material was the same in all the dwellings. After normalizing of the weighted radium concentration to 370 Bq per kg material, and of the ventilation rate to 1 air change per hour, the concentrations of radon and daughter products should be close to 740 Bq/m^3 and 220 Bq/m^3 respectively in the ground floor dwelling in a multi-family home in the above group. These values are higher than the corresponding values for single-family houses, 190 - 440 Bq/m^3 and 70 - 190 Bq/m^3 air respectively roughly in proportion to the amount of material. Our measurements therefore support the view that the radon concentration in dwellings near the ground can not be assigned solely to the building materials.

Single-family houses with aerated concrete based on alum shale, groups Nos. 5 and 6

In two of the groups of single-family houses, aerated concrete based on alum shale was included in the building material. In group No. 5 the walls of the cellars and in group No. 6 the outer and the inner walls of the dwellings were built of this material.

In group No. 5 the doors between the cellars and the dwelling parts of the houses were closed in about half of the investigated houses. After normalizing to 370 Bq radium per kg building material in the cellars and to 1 air change per hour, a radon concentration of 170 $\pm$ 11 Bq/m^3 air and a radon daughter concentration of 82 $\pm$ 11 Bq/m^3 air were found. These values are of the same magnitude as the corresponding values for single-family houses without aerated concrete based on alum shale. When the doors between the cellar and the dwelling part were open the concentrations of radon and daughter products were about half of the above values in the closed cellars.

Lower values for the concentrations of radon and radon daughters for one air change per hour and normalized to 370 Bq radium per kg building material was found for group No. 6. That this value is lower than for the cellars in group No. 5 is due to the fact that the concentration of radon and radon daughters for one air change per hour refers to the averages of the whole house including the first floor level where the radon levels are lower since the ceilings do not contain any stone material. The radon leakage from the ground may also be less for a ground floor dwelling than for a cellar which is partly surrounded by filling materials and soil.

Discussion and Conclusions

The absorbed dose from the external radiation normalized to m = 1 according to equation (1) is, as expected, found to be lower than the calculated value 1.5 mGy/y (150 mrad/year). For the multi-family houses the values were about 1.0 mGy/y and for the single-family houses about half that value.

The concentration of radon and daughter products for the maximum concentration of radium-226 in the building material according to equation (1) and normalized to one air change per hour was in accordance with the calculations of Krisiuk et al., about 110 Bq/m^3 for radon in equilibrium with the daughter products for the multi-family houses. For the single-family houses the concentration of radon in equilibrium with the daughter products does not differ significantly from the values in the multi-family houses despite the fact that the amounts of stony building materials were less than in the multi-family houses. For the non-cellar rooms in single-family houses built mainly with aerated concrete based on alum shale the values were lower.

The 1975 Swedish Building Code requires ventilation rate of at least 0.5 air changes per hour. This investigation showed that the ventilation rate in Swedish dwellings built in the beginning of the 1970's averaged about 0.5 air changes per hour.

If equation (1) should be used to define an exemption limit for new houses, the humans would be exposed to a maximum of 1.0 mGy/y from the building material in future houses. For the exemption limit the average concentration of radon would be about 200 Bq/m^3 air in equilibrium with the daughter products with the current ventilation requirements in Sweden.

The concentration of radium-226 in the ballast material, used for the manufacture of concrete in the town Gävle, is insignificantly higher than the average for Sweden (Ref 6). If a great part of the future buildings in Sweden were to consist of multi-family houses built of concrete containing natural ballast material and single-family houses built on a basement slab of concrete, the country-wide average with existing requirements for the ventilation rates, could be expected to be about 40 Bq $^{222}Rn/m^3$ air in equilibrium with the daughter products.

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1 Bq/m^3 0.027 pCi/l
1 mGy/year = 100 mrad/y

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Table 1. The concentration of radon and daughter products. The averages comprise all of the investigated dwellings with the exception of closed bedrooms and one dwelling where the window had been open before the measurement.

The type of building	Group No.	Building materials in the outer walls	Ventilation system	Air exchanges per hour			Radon Bq/m³ I	Radon Bq/m³ II	Radon daughters Bq/m³ I	Radon daughters Bq/m³ II	External radiation[a)] absorbed dose in gonads, mGy/y
Multi-family houses	1	concrete	F	0.3-0.6		lowest	59	48	22	11	0.53
						average	140	140	70	52	0.80
						highest	590	780	260	310	1.20
	2	concrete+ aerated concrete (based on sand)	F	0.5-0.9		lowest	37	26	13	9	0.64
						average	89	85	31	23	0.78
						highest	150	140	48	41	0.91
	3	concrete+ aerated concrete (bases on sand and on alum shale)	F	0.4-0.8		lowest	74	93	22	11	0.58
						average	180	160	52	44	0.80
						highest	440	410	150	140	2.05
Single-family houses	4	faced brick (sandstone) wood construction stone wool	F	0.4-0.7		lowest	n		8		0.33
						average	56		22		0.56
						highest	140		38		0.80
	5	wood and stone wool the cellar of aerated concrete based on alum shale	F	0.4-0.8	With cellars (8)	lowest	n		4		0.17
						average	200		81		0.38
						highest	370		140		0.71
					Without cellars (1)	lowest	100		33		0.35
						average	100		37		0.38
						highest	100		41		0.42

Table 2

The activity concentrations in the building materials of the kind used at the time of building the houses in the investigation.
1 Bq/kg ≈ 0.027 pCi/g

Building materials	Density	Origin		^{232}Th Bq/kg	^{226}Ra Bq/kg	^{40}K Bq/kg	m a)
Ballast		local gravelpits	lowest	67	52	700	0.56
			average		56		0.61
			highest	93	56	740	0.66
Cement		all manufacturers in Sweden	lowest	33	22	22	0.19
			average		56		0.38
			highest	110	190	370	1.0
Concrete	2.3	see note b)	lowest	59	44	590	0.48
			average		52		0.55
			highest	89	67	670	0.66
Aerated concrete based on sand	0.5	all manufacturers in Sweden	lowest	15	14	260	0.16
			average		74		0.56
			highest	115	120	480	0.75
Aerated concrete based on alum shale c)	0.5	Hällabrottet d)		70	1300	780	4.0
		average weighted for the production			1900		5.5
		Falköping d)		70	2200	960	6.5
Light-weight aggregate	0.3	all manufacturers in Sweden	lowest	150	110	890	1.06
			average		140		1.21
			highest	190	190	1100	1.45
Blocks of light weight aggregate		no adequate sampling	average	96	89	700	0.76
Brick (clay)	1.5	no adequate sampling	average	130	96	930	0.94

a) $m = \frac{C_{Th}}{260} + \frac{C_{Ra}}{370} + \frac{C_K}{4.8 \times 10^3}$

b) Estimated from measured values for ballast material and cement assuming water:cement:ballast = 6:12:82

c) No production since 1975

d) Names of factories

Table 3. Absorbed dose rate in the gonads from the external gamma radiation from the building material related to $m = \frac{C_{Th}}{260} + \frac{C_{Ra}}{370} + \frac{C_k}{4.8 \times 10^3}$ The activity concentrations giving m are weighted for the weights of the various building materials of the dwelling.

3 a MULTI-FAMILY HOUSES:

Group No	Absorbed dose in the gonads a) mGy/y	m	Absorbed dose in the gonads for m = 1: in the dwelling mGy/y	Absorbed dose in the gonads for m = 1: the outdoor dose subtracted b) mGy/y
1	0.76 ± 0.13	0.6 ± 0.1	1.27 ± 0.30	0.97 ± 0.30
2	0.76 ± 0.05	0.6 ± 0.1	1.27 ± 0.23	0.97 ± 0.23
3	0.85 ± 0.06	0.7 ± 0.1	1.21 ± 0.19	0.91 ± 0.19
Average			1.25 ± 0.24	0.95 ± 0.24

a) The average of measurements in a bed, an easy chair and at a dining table, excl. the contribution from the cosmic radiation, 0.3 mGy/y.
b) An average outdoor dose rate of 0.3 mGy/y has been subtracted. The error in the subtracted outdoor dose is not included in the standard deviation, since the value used is a roughly estimated average outdoor dose rate in Sweden.

1 mGy/y = 100 mrad/y.

3 b SINGLE-FAMILY HOUSES:

Group No	Absorbed dose a) in the gonads mGy/y	m	Abserbed dose b) in the gonads for m = 1 mGy/y
5 cellars	individually calculated because of various materials in the individual houses.		0.62 ± 0.12
6 ground floor	1.50 ± 0.06	2.0 ± 0.5	0.53 ± 0.17
7 ground floor	0.49 ± 0.03	0.6 ± 0.1	0.42 ± 0.10

a) The average of measurements in a bed, an easy chair and at a dining table excl. the contribution from the cosmic radiation, 0.3 mGy/y.

b) The error in the subtracted outdoor dose is not included in the standard deviation, since the value used is a roughly estimated average outdoor dose rate in Sweden.

Table 4. The concentration of radon and radon daughters calculated for 1 air exchange per hour and related to $m_{Ra} = \frac{C_{Ra}}{370}$. The concentration of radium is weighted for the weights of the various building materials in the dwelling.

4 a. MULTI-FAMILY HOUSES:

Group No	Radon Bq/m³	Radondaughters Bq/m³	m_{Ra}	For $m_{Ra} = 1$ Radon Bq/m³	For $m_{Ra} = 1$ Radondaughters Bq/m³
1	41 ± 15	15 ± 6	0,2 ± 0,05	200 ± 41	74 ± 33
2	56 ± 15	17 ± 6	0,2 ± 0.05	280 ± 100	89 ± 37
3	96 ± 22	24 ± 6	0,3 ± 0,05	320 ± 110	81 ± 26
Average				270 ± 90[a]	81 ± 33[a]

4 b. SINGLE-FAMILY HOUSES:

Group No	Radon Bq/m³	Radondaughters Bq/m³	m_{Ra}	For $m_{Ra} = 1$ Radon Bq/m³	For $m_{Ra} = 1$ Radondaughters Bq/m³
4	26 ± 11	11 ± 5	0,2 ± 0,05	130 ± 66	56 ± 29
The cellars in No 5[b]	220 ± 130	107 ± 67	0,9 – 2,1	170 ± 11	82 ± 11
6	89 ± 37	37 ± 15	1,6 ± 0,3	59 ± 30	22 ± 11
7	78 ± 15	33 ± 11	0,2 ± 0,05	410 ± 150	170 ± 70

1 Bq/m³ ≈ 0,027 pCi/l.

a) One standard deviation without the error of the calibration constant.

b) The doors between the cellars and the ground floor rooms were closed.