Radiological characterization and dose assessment of Israeli concrete

Survey 2011 - 2016

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Contents

List of tables

List of figures

Summary

The aim of this study is to investigate the radiation impact from concrete used in building construction with fly ash compared against concrete without. For this purpose a total of five surveys are performed each involving four to five different concrete mixtures to determine the activity concentrations and the radon exhalation, followed by an assessment of the radiation dose when these concrete mixtures are used in regular dwellings. The concrete mixtures include a reference concrete without fly ash, followed by three or four other samples with fly ash derived from various coal sources. Experiments were carried out in compliance with the Dutch standards NEN 5697 and NEN-ISO 11665-9. The results demonstrate a total annual effective dose from concrete with no fly ash ranging from 0.76 to 0.82 mSv, while for concrete with fly ash the annual dose ranges from 0.70 up to 0.96 mSv.

Introduction

Fly ash – a by-product of burning pulverized coal in an electrical generating station – is at present widely used as a supplementary cementitious material in the production of cement and concrete. In cement fly ash is found as a partial replacement of clinker, while in concrete it is used as a replacement of sand and cement. Its potential as a supplementary material has been known almost since the start of the last century and offers significant benefits. However, as for all materials of mineral origin fly ash is a source for natural radioactivity, due to the presence of ^{226}Ra (radium), ^{232}Th (thorium) and ^{40}K (potassium). These radionuclides are a source for gamma-emitting (decay) products in the building materials resulting in an external radiation dose to the inhabitant. Furthermore, the presence of ^{226}Ra also provides a source of radon and contributes to the radon exposure.

The aim of this study is to investigate the radiation impact from concrete used in building construction with fly ash additive compared against concrete without. For this purpose a total of five surveys are performed each involving four to five different concrete mixtures. Each survey includes a single mixture without fly ash followed by three or four mixtures with fly ash. The concrete mixtures are tested in the laboratory to determine its radiological properties, followed by a radiological assessment to determine the radiation exposure from external radiation and radon for a typical room construction.

This report first describes the methods to determine the radiological properties of the concrete mixtures and the modelling techniques used in the assessment. The method description is followed by an overview of the results and ends with a summary of the main findings.

1 Testing and modelling methods

1.1 Experimental testing

As part of the presented work a total of 21 different concrete mixtures are studied to determine the activity concentrations from ^{226}Ra, ^{232}Th (^{228}Ra and ^{228}Th) and ^{40}K, and the radon exhalation rate. Each set contains one mixture without fly ash followed by three to four mixtures with fly ash of different origin. For this purpose concrete samples were provided in dual. The activity content was determined in three identical samples (0.1×0.1×0.1 m^3) for each concrete mixture. These samples were crushed by the laboratory prior to the measurements with a particle size smaller than 0.1 cm. The radon exhalation rate was measured in a single test using a separate set of three identical samples (0.1×0.1×0.2 m^3).

1.1.1 Activity concentrations

The natural radioactivity concentrations (a in $Bq \cdot m^{-3}$) of the specimens are determined according to a standard method published under NEN 5697[3,4]. According to this method the density dependent photo peak efficiencies are determined for the gamma-ray energies 352 keV (^{214}Pb, parent ^{226}Ra), 583 keV (^{208}Tl, parent ^{228}Th), 911 keV (^{228}Ac, parent ^{228}Ra) and 1,461 keV (^{40}K). Four calibration standards are assembled with increasing densities. The materials used are stearic acid, starch, gypsum and quartz sand, homogeneously mixed with certified amounts of ^{238}U and ^{232}Th, in equilibrium with their daughter nuclides, and ^{40}K. The standards are placed into Marinelli beakers with a volume of about 1 liter, weighted and closed radon-tight. To obtain secular equilibrium, a waiting time of at least three weeks is taken into account before counting the samples. All samples are counted using an HPGe detector in a low-background facility. The samples of the material are analysed in an identical way as the calibration standards with respect to geometry, waiting time and radon-tightness of the beaker. The photo-peak efficiencies of the samples are deduced from the efficiency curves of the standard samples by interpolation. The results are expressed per unit of dry weight.

1.1.2 Radon exhalation rate

The natural free radon exhalation rate (ER_{Rn} in $Bq \cdot (kg \cdot s)^{-1}$) of the concrete samples is determined according to the standard method published under NEN 5699[5]. Determination of the radon exhalation rate is based on a continuous ventilation of an exhalation chamber with material sample. On the outlet side of the chamber the ^{222}Rn from the material sample is collected and subsequently quantified using liquid scintillation counting. For this purpose an exhalation chamber with an approximate volume of around 36 liter is required. A constant flow of radon-free nitrogen gas of known humidity is passed through the chamber. The relative humidity of the nitrogen flow is regulated within the full range of 0 to 100% by means of a controlled mixing of dry and water–saturated nitrogen gas (Figure 1). After a given time (normally within 3 h) a steady-state concentration is reached and the experiment can be started. The out coming flow is guided through two U-shaped tubes for a period of 10 to 30 minutes. The first tube contains KOH tablets to dry the gas flow; the second tube contains 4 g of silica gel and is cooled with liquid nitrogen to trap the radon. After absorption, the tube with silica gel is warmed and subsequently the content is poured into a counting vial containing toluene-based scintillation liquid. During this process no loss of ^{222}Rn was observed.

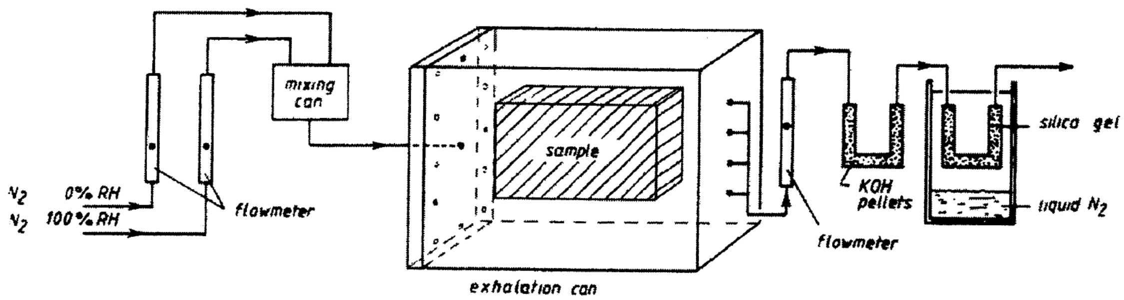

Figure 1 Schematic view of the measuring arrangement for determination of the radon exhalation rate according to NEN 5699.

Radioactive equilibrium in the counting vial is attained after around 3 h, and a further 13 h is required before the radon progeny in the vial is in equilibrium. Subsequently, a recording of the spectrum is performed using a liquid scintillation spectrometer. For an optimal count rate the window settings are set from 110 to 600 keV. Under these conditions a counting efficiency of approximately 2.8 $c \cdot s^{-1} \cdot Bq^{-1}$ can be reached.

The samples are conditioned at a temperature of 20 ± 2°C and a relative humidity of 50 ± 5%. Equilibrium is achieved when the mass of the sample over a period of seven days deviates by less than 0.07% from the value determined during the previous measurement. For fresh concrete a minimum curing period of at least 28 days is required.

1.2 Modelling approach

1.2.1 Indoor radon and radon progeny concentration

In this work a Computational Fluid Dynamics (CFD) model is used to simulate the concentration of radon and radon progeny concentrations (C) in a standard Israeli shelter room (which is made of massive concrete). The dispersion of air and radon (progeny) is computed using the fundamental flow equations for gas and aerosols, which enables detailed simulation of the three-dimensional flow structures from ventilation and buoyancy. Algorithms are incorporated and coupled with the CFD model to take account of all relevant physical processes. These include the formation and attachment of the progeny products to aerosol particles as well as the dispersion and deposition of the radioactive aerosols. Further details on the modelling technique are described by De With and De Jong[6]. The computations are performed using the radon exhalation rate (ER_{Rn}) as measured according to NEN 5699 and applied to all wall surfaces.

1.2.2 Dose modelling

External dose

The external exposure component of the effective dose (E_{Ex}) from the building materials in the room is calculated according to the method described by De Jong and Van Dijk[7,8]. The method is based on a standard room geometry of 5 by 4 m and 2.8 m in height as defined by Koblinger[9]. Each construction part (i.e. floor, walls and ceiling) is made of 20 cm thick concrete and no doors or windows. Correction factors are deduced for alternative situations. The absorbed dose rate in air (unit: Gy·h^{-1}) in a particular room is than calculated according to:

$$\dot{D}_{air} = \left\{ \sum_{i=1}^{6} \left[F_{dose} \cdot F_1 \cdot F_2 \cdot F_3 \cdot \dots \cdot F_n \right]_i \right\} F_{zoning} \cdot F_{adjac} \qquad (1)$$

in which i is the index for a construction part, F_1 to F_n are correction factors for each construction part i, where n=4. F_{zoning} is a correction factor which takes internal zoning of the construction into account, and F_{adjac} is the contribution from adjacent floors and dwellings. F_{dose} is the so-called dose factor, defined as: $F_{dose,i}=k_1 \cdot a_{1,i}+k_2 \cdot a_{2,i}+k_3 \cdot a_{3,i}$. In this equation k_1, k_2 and k_3 represent the specific absorbed dose rates, and $a_{1,i}$, $a_{2,i}$ and $a_{3,i}$ the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K of construction part i (Bq·kg^{-1}), respectively. The activity concentrations are obtained experimentally according to NEN 5697. The values of the specific absorbed dose rate depend amongst others on the thickness, density and dimensions of the various construction parts and are selected as 0.90, 1.10 and 0.08 nGy·h^{-1} per Bq·kg^{-1} respectively[10]. The absorbed dose is multiplied with a conversion factor of 0.7 Sv·Gy^{-1} to obtain the effective dose E_{Ex}.

Radon dose

The radon component of the effective dose (E_{Rn}) in mSv per year is computed according to UNSCEAR[11]:

$$E_{Rn} = DCC_{Rn} \cdot EEC_{Rn} \cdot t_{Rn}, \qquad (2)$$

where DCC_{Rn} is the dose conversion coefficient of 9 nSv·h^{-1} per Bq·m^{-3}, EEC_{Rn} the equilibrium equivalent ^{222}Rn concentration and t_{Rn} is the hours per year spent indoors. The EEC_{Rn} is calculated as $EEC_{Rn} = 0.105 C_1 + 0.515 C_2 + 0.380 C_3$, where C_1, C_2 and C_3 are the activity concentrations in the indoor environment of ^{218}Po, ^{214}Pb and ^{214}Bi (Bq·m^{-3}), respectively. These concentrations are obtained from the CFD computations.

2 Results

2.1 Experimental results

2.1.1 Activity concentrations

A gamma-spectrometric analysis on the radioactivity concentrations of the gamma-ray emitting radionuclides is carried out in three samples of each of the sixteen concrete mixtures. The results from these measurements are presented in Table 1. The results demonstrate a ^{226}Ra ranging between 29 and 41 $Bq \cdot kg^{-1}$ with elevated ^{226}Ra concentrations for the mixtures with fly ash. The concentrations ^{228}Ra and ^{228}Th, which are both part of the ^{232}Th series are in the order of 4 to 17 $Bq \cdot kg^{-1}$. In each mixture the nuclide concentration of ^{228}Ra and ^{228}Th is broadly similar indicating that the thorium decay series is in secular equilibrium. The concentrations for ^{40}K range between 40 to 70 $Bq \cdot kg^{-1}$ and also here the activity in the mixtures with fly ash is higher.

2.1.2 Radon exhalation rate

To obtain the ^{222}Rn exhalation rate a single test is carried out for each concrete mixture according to the Dutch standard NEN 5699[5]. The results from these experiments are presented in Table 1 and show a radon exhalation rate (ER_{Rn}) of approximately 4.5 to 8.4 $\mu Bq \cdot (kg \cdot s)^{-1}$. The radon exhalation from the mixtures with fly ash is in most cases higher. However, in the surveys from 2011-2012 to 2015 there are five cases where the exhalation is lower than the concrete without fly ash. This reduction occurs despite the higher ^{226}Ra concentrations in the mixtures with fly ash. Table 1 also shows the radon exhalation factor (ER_f), which is defined as the ratio between the amount of radon released to the environment and the amount that is generated within the material, and represents the percentage of radon that is released from the concrete. The factor is computed as $ER_f = ER_{Rn}/(C_{Ra\text{-}226} \cdot \lambda_{Rn})$, where ER_{Rn} is the exhalation rate in $Bq \cdot (kg \cdot s)^{-1}$, $C_{Ra\text{-}226}$ the radium concentration in $Bq \cdot kg^{-1}$ and λ_{Rn} the radon decay constant. With the exception of two concrete mixtures the ER_f is lower for all mixtures with fly ash and demonstrates that the percentage of radon released is reduced when fly ash is added.

Table 1 Activity concentrations (*a*) with its standard uncertainty (±1 SD) expressed in Bq·kg^{-1}, and radon exhalation rate (*ER*) with its standard uncertainty (±1 SD) expressed in µBq·s^{-1} and µBq·(kg·s)$^{-1}$. The first mixture in each survey is without fly ash. Fly ash content in 2011-2012's mixtures is 120 kg/m^3 and 100 kg/m^3 in 2013's mixtures and on.

Survey 2011-2012	Activity concentrations				Radon exhalation		
	$a_{Ra\text{-}226}$	$a_{Ra\text{-}228}$	$a_{Th\text{-}228}$	$a_{K\text{-}40}$	ER_{Rn}		ER_f
Sample	(Bq·kg^{-1})	(Bq·kg^{-1})	(Bq·kg^{-1})	(Bq·kg^{-1})	(µBq·s^{-1})	(µBq·(kg·s)$^{-1}$)	(%)
7013-A	34.0±2.0	6.0±0.4	6.1±0.5	48.0±3.0			
7013-B	35.0±2.0	6.5±0.4	6.7±0.5	49.0±3.0			
7013-C	35.0±4.0	5.8±0.5	5.8±0.6	46.0±2.0			
7013	34.7±2.7	6.1±0.4	6.2±0.5	47.7±2.7	90.3±5.0	6.3±0.3	8.7
7014-13	36.0±2.0	8.4±0.5	8.7±0.7	75.0±3.0			
7014-14	33.0±3.0	7.9±0.8	8.5±0.8	68.0±5.0			
7014-15	37.0±2.0	8.5±0.5	9.5±0.7	78.0±3.0			
7014	35.3±2.3	8.3±0.6	8.9±0.7	73.7±3.7	87.2±19.8	6.1±1.4	8.2
7015-13	36.0±4.0	10.2±0.7	9.9±0.9	48.0±2.0			
7015-14	37.0±4.0	10.0±0.7	10.9±0.9	50.0±2.0			
7015-15	37.0±2.0	9.8±0.5	10.9±0.7	49.0±3.0			
7015	36.7±3.3	10.0±0.6	10.6±0.8	49.0±2.3	103.7±2.1	7.2±0.1	9.4
7017-16	41.0±2.0	9.8±0.5	9.8±0.7	74.0±3.0			
7017-17	40.0±2.0	8.8±0.5	8.3±0.6	73.0±3.0			
7017-18	40.0±2.0	8.9±0.7	9.8±1.0	73.0±3.0			
7017	40.3±2.7	9.2±0.6	9.3±0.8	73.3±3.0	84.4±15.9	5.9±1.1	6.9
7019-20	36.0±2.0	9.3±0.5	9.2±0.6	55.0±3.0			
7019-21	38.0±2.0	8.1±0.6	9.5±1.0	59.0±3.0			
7019-22	47.0±4.0	11.3±0.8	10.9±1.0	72.0±3.0			
7019	40.3±2.7	9.6±0.6	9.9±0.6	62.0±3.0	84.4±10.2	5.8±0.7	6.9

Survey 2013 Sample	Activity concentrations a_{Ra-226} (Bq·kg^{-1})	a_{Ra-228} (Bq·kg^{-1})	a_{Th-228} (Bq·kg^{-1})	a_{K-40} (Bq·kg^{-1})	Radon exhalation ER_{Rn} (µBq·s^{-1})	ER_{Rn} (µBq·(kg·s)$^{-1}$)	ER_f (%)
2872-A	33.6±2.0	5.0±0.5	4.2±0.4	43.3±1.9			
2872-B	33.0±3.0	5.0±0.6	4.9±0.5	42.0±4.0			
2872-C	31.8±0.8	5.5±0.4	5.1±0.3	41.7±1.5			
2872	32.8±1.9	5.2±0.5	4.7±0.4	42.3±2.5	106.8±5.6	7.3±0.4	10.6
5850-A	39.0±3.0	14.8±0.9	15.2±1.0	43.0±5.0			
5850-B	39.0±3.0	15.1±0.7	14.4±0.7	50.0±3.0			
5850-C	40.0±3.0	15.6±1.0	14.8±0.8	48.0±3.0			
5850	39.3±3.0	15.2±0.9	14.8±0.8	47.0±3.7	121.5±14.4	8.4±1.0	10.2
5852-A	36.0±3.0	7.8±0.5	7.7±0.5	62.0±4.0			
5852-B	34.6±0.9	8.2±0.6	8.4±0.4	57.8±1.8			
5852-C	35.0±3.0	8.1±0.5	8.3±0.6	51.0±6.0			
5852	35.2±2.3	8.0±0.5	8.1±0.5	56.9±3.9	103.8±8.9	7.2±0.6	9.8
7699-A	36.0±3.0	7.6±0.5	7.5±0.6	54.4±1.6			
7699-B	36.0±3.0	8.2±0.8	8.4±0.8	61.0±6.0			
7699-C	37.0±3.0	7.6±0.6	7.4±0.4	53.0±3.0			
7699	36.3±3.0	7.8±0.6	7.8±0.6	56.1±3.5	121.3±13.7	8.3±0.9	10.9

Survey 2014 Sample	Activity concentrations a_{Ra-226} (Bq·kg^{-1})	a_{Ra-228} (Bq·kg^{-1})	a_{Th-228} (Bq·kg^{-1})	a_{K-40} (Bq·kg^{-1})	Radon exhalation ER_{Rn} (µBq·s^{-1})	ER_{Rn} (µBq·(kg·s)$^{-1}$)	ER_f (%)
7335-A	29.0±3.0	6.0±0.7	6.1±0.6	49.0±5.0			
7335-B	29.0±0.8	5.5±0.5	5.3±0.3	46.0±1.5			
7335-C	30.1±1.9	5.9±0.4	5.3±0.4	49.0±3.0			
7335	29.4±1.9	5.8±0.5	5.6±0.4	48.0±3.2	100.9±3.5	6.9±0.2	11.3
7336-A	35.7±2.0	11.5±0.8	11.5±0.6	63.0±3.0			
7336-B	38.0±3.0	12.1±0.7	11.3±0.7	62.0±1.7			
7336-C	36.2±1.8	11.9±0.6	13.1±0.6	70.0±3.0			
7336	36.6±2.3	11.8±0.7	12.0±0.6	65.0±2.6	103.9±1.9	7.2±0.1	9.3
7337-A	35.7±0.9	9.5±0.6	9.0±0.5	67.5±1.9			
7337-B	36.0±3.0	9.0±0.9	8.6±0.8	68.0±6.0			
7337-C	33.5±2.0	9.6±0.7	9.6±0.6	75.0±3.0			
7337	35.1±2.0	9.4±0.7	9.1±0.6	70.2±3.6	105.8±7.8	7.3±0.5	9.9
7338-A	42.0±3.0	18.1±1.1	17.1±0.9	60.0±3.0			
7338-B	41.0±3.0	15.5±0.9	15.5±1.0	52.0±1.6			
7338-C	41.0±4.0	16.0±1.4	15.8±1.4	55.0±5.0			
7338	41.3±3.3	16.5±1.1	16.1±1.1	55.7±3.2	106.4±6.2	7.3±0.4	8.4

Survey 2015	Activity concentrations				Radon exhalation		
	a_{Ra-226}	a_{Ra-228}	a_{Th-228}	a_{K-40}	ER_{Rn}		ER_f
Sample	(Bq·kg^{-1})	(Bq·kg^{-1})	(Bq·kg^{-1})	(Bq·kg^{-1})	(μBq·s^{-1})	(μBq·(kg·s)$^{-1}$)	(%)
9053-4	31.0±3.0	6.9±0.7	6.4±0.7	50.0±5.0			
9053-5	30.0±0.8	5.4±0.4	5.8±0.3	40.8±1.4			
9053-6	31.4±1.7	5.1±0.4	5.5±0.3	44.0±3.0			
9053	30.8±1.8	5.8±0.5	5.9±0.4	44.9±3.1	93.9±3.3	6.2±0.2	9.6
9054-4	37.0±3.0	13.0±0.8	12.5±0.9	49.0±1.4			
9054-5	39.0±3.0	14.1±0.9	13.7±0.8	53.0±3.0			
9054-6	37.0±3.0	13.8±1.2	13.6±1.3	50.0±5.0			
9054	37.7±3.0	13.6±1.0	13.3±1.0	50.7±3.1	98.2±11.0	6.5±0.7	8.2
9055-4	36.8±2.0	7.0±0.4	6.0±0.4	50.0±3.0			
9055-5	39.0±3.0	6.8±0.5	6.9±0.4	52.0±4.0			
9055-6	36.1±0.9	6.2±0.5	6.3±0.3	48.4±1.5			
9055	37.3±2.0	6.7±0.5	6.4±0.4	50.1±2.8	68.6±3.7	4.5±0.2	5.8
9056-1	41.0±3.0	9.6±0.6	9.6±0.7	33.1±1.2			
9056-2	41.0±3.0	9.6±0.7	9.7±0.6	37.3±1.8			
9056-3	41.0±3.0	10.7±0.6	11.1±0.6	40.6±2.0			
9056	41.0±3.0	10.0±0.6	10.1±0.6	37.0±1.7	109.8±3.7	7.3±0.2	8.5

Survey 2016	Activity concentrations				Radon exhalation		
	a_{Ra-226}	a_{Ra-228}	a_{Th-228}	a_{K-40}	ER_{Rn}		ER_f
Sample	(Bq·kg^{-1})	(Bq·kg^{-1})	(Bq·kg^{-1})	(Bq·kg^{-1})	(μBq·s^{-1})	(μBq·(kg·s)$^{-1}$)	(%)
2995-A	34.0±3.0	4.5±0.4	4.6±0.4	41.0±1.2			
2995-B	33.0±3.0	4.3±0.4	4.3±0.3	41.0±3.0			
2995-C	32.4±0.8	4.3±0.3	4.5±0.4	38.1±1.4			
2995	33.1±2.3	4.4±0.4	4.5±0.4	40.0±1.9	100.6±5.4	6.8±0.4	9.8
4645-A	35.1±1.9	5.9±0.5	6.4±0.4	64.0±3.0			
4645-B	36.0±3.0	6.8±0.7	6.5±0.7	61.0±6.0			
4645-C	35.3±1.8	7.1±0.5	6.7±0.4	68.0±3.0			
4645	35.5±2.2	6.6±0.6	6.5±0.5	64.3±4.0	113.1±5.3	7.6±0.4	10.3
4646-A	40.0±4.0	12.5±1.1	12.5±1.2	45.0±4.0			
4646-B	39.0±3.0	13.5±0.9	12.8±0.7	48.0±3.0			
4646-C	40.0±3.0	12.4±0.6	13.6±0.7	50.0±3.0			
4646	39.7±3.3	12.8±0.9	13.0±0.9	47.7±3.3	113.8±6.8	7.7±0.5	9.2
4647-A	36.1±0.9	8.5±0.6	8.1±0.4	62.5±1.9			
4647-B	37.0±3.0	6.9±0.7	6.9±0.7	62.0±6.0			
4647-C	36.2±0.9	7.9±0.5	7.3±0.4	59.0±1.8			
4647	36.4±1.6	7.8±0.6	7.4±0.5	61.2±3.2	107.6±7.3	7.3±0.5	9.6

2.2 Modelling results

Based on the experimental findings reported in Section 2.1 the dose from external radiation and radon is computed using the methods described in Section 1.2. For the computation a room with 20 cm thick concrete walls, floor and ceiling is assumed. The time spent indoors is taken as 7000 h per year, which corresponds with 80% of the total time[11].

2.2.1 Indoor radon and radon progeny concentration

CFD calculations are performed for the different concrete mixtures according to Section 1.2.1. The modelling is based on a ventilated room with an air exchange rate of 0.5 h^{-1}. The radon exhalation applied at the wall is obtained from the experimental data presented in Table 1. For this purpose the radon exhalation rate is corrected with a correction factor of 0.79[12]. This correction is required as the radon exhalation is measured from all six surfaces of the sample, while under real conditions the exhalation is one-dimensional and will only take place from the two outer surfaces of the building element. The background concentration of radon is 10 $Bq\cdot m^{-3}$ with an equilibrium factor between radon and its progeny of 0.4[11]. Further model assumptions are as follows;

Room dimensions	: length 3 m; width 3 m; height 2.7 m
Inner area (A_{Ro})	: 50.4 m^2
Volume (V)	: 24.3 m^3
Window (A_{Wi})	: 1.2 x 1.2 m^2 (1.4 m^2)
Door (A_{Do})	: 2.0 x 0.8 m^2 (1.6 m^2)
Area concrete (A_{Co})	: 47.4 m^2
Concrete thickness (L)	: 0.2 m
Concrete density (ρ)	: 2300 $kg\cdot m^{-3}$
Furniture	: Unfurnished room

The computed concentrations for radon and its progeny (C) in the reference room are presented in Table 2. The radon concentrations are in the order of 20 to 30 $Bq\cdot m^{-3}$, and the progeny concentrations are reduced to around 5 $Bq\cdot m^{-3}$ for ^{214}Bi due to natural ventilation and deposition of the radon progeny. The concentrations include the radon background of 10 $Bq\cdot m^{-3}$ and its background progeny.

Table 2 Radon and radon progeny (C) in Bq·m^{-3} and annual dose from radiation exposure (E) in mSv per year, with its standard uncertainty (±1 SD), where E_{tot}=E_{Ex}+E_{Rn}. The reported uncertainty is based on the experimental uncertainty reported in Table 1.

Survey 2011-2012	Radon (progeny)				Annual effective dose		
	$C_{Rn\text{-}222}$	$C_{Po\text{-}218}$	$C_{Pb\text{-}214}$	$C_{Bi\text{-}214}$	E_{Ex}	E_{Rn}	E_{Tot}
Sample	(Bq·m^{-3})	(Bq·m^{-3})	(Bq·m^{-3})	(Bq·m^{-3})	(mSv·yr^{-1})	(mSv·yr^{-1})	(mSv·yr^{-1})
7013	26.1±0.9	22.2±0.7	9.5±0.3	4.9±0.1	0.20±0.01	0.57±0.02	0.78±0.02
7014	25.4±3.5	21.6±2.9	9.2±1.1	4.8±0.5	0.23±0.01	0.56±0.07	0.79±0.07
7015	28.5±0.4	24.2±0.3	10.3±0.1	5.3±0.1	0.24±0.02	0.62±0.01	0.86±0.02
7017	24.9±2.8	21.2±2.3	9.1±0.9	4.7±0.4	0.26±0.01	0.55±0.06	0.80±0.06
7019	24.8±1.8	21.2±1.5	9.1±0.6	4.7±0.3	0.25±0.02	0.55±0.04	0.80±0.04

Survey 2013	Radon (progeny)				Annual effective dose		
	$C_{Rn\text{-}222}$	$C_{Po\text{-}218}$	$C_{Pb\text{-}214}$	$C_{Bi\text{-}214}$	E_{Ex}	E_{Rn}	E_{Tot}
Sample	(Bq·m^{-3})	(Bq·m^{-3})	(Bq·m^{-3})	(Bq·m^{-3})	(mSv·yr^{-1})	(mSv·yr^{-1})	(mSv·yr^{-1})
2872	29.0±1.0	24.7±0.8	10.4±0.3	5.4±0.2	0.19±0.01	0.63±0.02	0.82±0.02
5850	31.7±2.6	26.9±2.1	11.3±0.8	5.8±0.4	0.27±0.01	0.68±0.05	0.96±0.05
5852	28.5±1.6	24.2±1.3	10.3±0.5	5.3±0.2	0.22±0.01	0.62±0.03	0.84±0.03
7699	31.6±2.4	26.8±2.0	11.3±0.8	5.8±0.4	0.22±0.01	0.68±0.05	0.91±0.05

Survey 2014	Radon (progeny)				Annual effective dose		
	$C_{Rn\text{-}222}$	$C_{Po\text{-}218}$	$C_{Pb\text{-}214}$	$C_{Bi\text{-}214}$	E_{Ex}	E_{Rn}	E_{Tot}
Sample	(Bq·m^{-3})	(Bq·m^{-3})	(Bq·m^{-3})	(Bq·m^{-3})	(mSv·yr^{-1})	(mSv·yr^{-1})	(mSv·yr^{-1})
7335	27.9±0.6	23.7±0.5	10.1±0.2	5.2±0.1	0.18±0.01	0.61±0.01	0.79±0.02
7336	28.4±0.3	24.2±0.3	10.2±0.1	5.3±0.1	0.25±0.01	0.62±0.01	0.87±0.01
7337	28.7±1.4	24.4±1.2	10.3±0.5	5.3±0.2	0.23±0.01	0.63±0.03	0.86±0.03
7338	28.9±1.1	24.5±0.9	10.4±0.4	5.3±0.2	0.29±0.02	0.63±0.02	0.92±0.03

Survey 2015	Radon (progeny)				Annual effective dose		
	$C_{Rn\text{-}222}$	$C_{Po\text{-}218}$	$C_{Pb\text{-}214}$	$C_{Bi\text{-}214}$	E_{Ex}	E_{Rn}	E_{Tot}
Sample	(Bq·m^{-3})	(Bq·m^{-3})	(Bq·m^{-3})	(Bq·m^{-3})	(mSv·yr^{-1})	(mSv·yr^{-1})	(mSv·yr^{-1})
9053	26.0±0.6	22.2±0.5	9.5±0.2	4.9±0.1	0.18±0.01	0.57±0.01	0.76±0.01
9054	26.7±1.9	22.8±1.6	9.7±0.6	5.0±0.3	0.26±0.01	0.58±0.04	0.84±0.04
9055	21.6±0.6	18.4±0.5	8.0±0.2	4.2±0.1	0.22±0.01	0.48±0.01	0.70±0.02
9056	28.9±0.6	24.6±0.5	10.4±0.2	5.3±0.1	0.25±0.02	0.63±0.01	0.88±0.02

Table 2 continued.

Survey 2016 Sample	Radon (progeny) C_{Rn-222} (Bq·m^{-3})	C_{Po-218} (Bq·m^{-3})	C_{Pb-214} (Bq·m^{-3})	C_{Bi-214} (Bq·m^{-3})	Annual effective dose E_{Ex} (mSv·yr^{-1})	E_{Rn} (mSv·yr^{-1})	E_{Tot} (mSv·yr^{-1})
2995	27.4±0.9	23.3±0.8	9.9±0.3	5.1±0.1	0.19±0.01	0.60±0.02	0.78±0.02
4645	29.5±0.9	25.1±0.8	10.6±0.3	5.4±0.1	0.22±0.01	0.64±0.02	0.86±0.02
4646	29.6±1.2	25.2±1.0	10.6±0.4	5.5±0.2	0.26±0.02	0.64±0.02	0.91±0.03
4647	28.8±1.3	24.5±1.1	10.4±0.4	5.3±0.2	0.23±0.01	0.63±0.03	0.85±0.03

2.2.2 Dose modelling

External dose

Following the above described results, a dose assessment for external radiation and radon exposure is performed for the different concrete mixtures. For this purpose the concentrations ^{228}Ra and ^{228}Th (*a*) reported in Table 1 are averaged to obtain an estimated ^{232}Th concentration in the building material. The annual effective dose (E_{Ex}) from external radiation is shown in Table 2. The results show a dose from external radiation of around 0.18 to 0.29 mSv per year. The external dose is higher for the samples with fly ash due to its increase in activity concentrations.

Radon dose

The annual effective dose from radon (E_{Rn}) is also shown in Table 2 and is around 0.5 to 0.7 mSv. This includes exposure to radon's background concentration of 10 Bq·m^{-3}. As a result the dose from radon is considerable higher than the dose from external radiation. However, the variation in annual radon dose from the different concrete mixtures is limited and ranges between 0.48 mSv and 0.68 mSv. By accumulating both doses the total dose from external radiation and radon is around 0.8 mSv with a maximum of over 0.9 mSv per year for certain types of concrete with fly ash.

3 Conclusions

A total of five surveys each involving four to five different concrete mixtures have been studied to determine the activity concentrations and the radon exhalation, followed by a calculation of the annual effective dose when these concrete mixtures are used in regular dwellings. The concrete mixtures include a reference concrete without fly ash, followed by three or four other samples with fly ash derived from various coal sources. Experiments were carried out in compliance with the Dutch standards NEN 5697 and NEN-ISO 11665-9.

The results demonstrate an increase in the total effective dose from around 0.8 mSv with no fly ash and 0.70 mSv to 0.96 mSv when fly ash is used. The increase is by and large a result of increased exposure to external radiation, while the annual radon dose remains broadly similar. The annual radon dose remains similar as the addition of fly ash reduces the radon exhalation factor in nearly all cases and sometimes even reduces the radon exhalation rate despite the higher ^{226}Ra concentration.

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