10 נובמבר, 2017
מנ - 39557

חישוב מנת הקרינה הנתרמת ע"י האפר כתוסף לבטון במודל ההולנדי

מאז 2012 במשך 5 שנים בצעה מעבדת NRG בהולנד (מעבדה מתמחה המשמשת לרגולציה) עבור המנהלת בדיקות קרינה בדוגמאות בטון-אפר ישראלי מייצגות, במקביל לבדיקות השוטפות הנערכות מדי שנה במעבדות שורק ובן גוריון. מטרת הבדיקות המקבילות היתה להעמיד במבחן גורם בינלאומי בעל מוניטין את שיטת הבדיקה שפותחה בישראל לשפיעת רדון ממוצרי בנייה ואת מודל הערכת מנת הקרינה המהווה בסיס לחישובי ת.י. 5098 – תכולת יסודות רדיואקטיביים טבעיים במוצרי בנייה, וכן לשמש כמעבדת ביקורת Round Robin למדידת תכולת היסודות הרדיואקטיביים במעבדת שורק בשיטה המקובלת בעולם.

ממצאי המדידות (השוואה בין מעבדתית מצורפת) מצביעים על שני מאפיינים עקביים:

- ערכי ריכוזי היסודות הרדיואקטיביים וקצב שפיעת הרדון המתקבלים ב-NRG גבוהים במקצת מאשר מקביליהם בשורק. ההבדלים בממצאי הרדון מיוחסים להבדל בשיטות המדידה.
- תרומת אפר הפחם למנת הקרינה המתקבלת בהפרש ממצאי הבטון עם אפר פחם ובלעדיו דומה בשתי המעבדות.

יצוין כי בעוד השיטה הישראלית לא נשפטה ע"י האקדמיה בעולם, השיטה ההולנדית מתועדת זה שנים רבות בפרסומים בעיתונות המקצועית הבינלאומית (מצורף האחרון שבהם המסכם את ממצאי הבדיקות שנערכו בהזמנת המנהלת) ולכן ממצאיה מהווים מעין אישור עקיף לאיכות ממצאי הבדיקות בארץ (מצורף מאמר השוואתי המאשש מסקנה זו).

הבדיקות שבוצעו במעבדת NRG, הן מבחינת היקפן (21 סדרות, 5 מתוכן ללא אפר פחם) והן מבחינת מגוון המקורות הפחם (16 מקורות, המייצגים את מרבית אפר הפחם מדי שנה), משקפות את אפר הפחם שנעשה בו שימוש כתוסף לבטון בשנים אלה, וככל שניתן להעריך את הרכב סל הפחם העתידי, גם את אפר הפחם שיעשה בו שימוש בשנים הבאות.

כאמור לעיל מטרת הבדיקות היתה להעמיד במבחן את תוצאות הבדיקות במעבדות בארץ על פי דרישות ת.י. 5098. ת.י. 5098 נועד לקבוע סף עליון מותר של ריכוזי יסודות במוצרי בנייה במקרה הקיצוני של חדר הבנוי בשש פאותיו ממוצר הבנייה הנבחן, כלומר ממ"ד במקרה השימוש בבטון, בשהייה של 7000 שעות בשנה (19 שעות ביממה). מקרה זה, המייצג מצב קיצוני של אדם המרותק מרבית שעות היממה למבנה נתון לכל אורך חייו, איננו משקף את מנת הקרינה לה נחשף אדם מייצג מהציבור.

לצורך הערכה ברמה הלאומית של מנת הקרינה מאפר פחם כתוסף לבטון לה נחשפת האוכלוסייה בישראל בכללה ואוכלוסיות משנה בתוכה, יש להשתמש במודל מורכב המתייחס לדירה מייצגת הכוללת חדרים בעלי אפיונים שונים – חדרי שינה, חדר מגורים, מטבח, שירותים ומסדרונות ולמשך שהייה נורמטיבי בכל אחד מהחדרים.

מודל התקן ההולנדי בנוי באופן מכליל המאפשר להעריך את מנות הקרינה בכל נקודה בדירה בהתייחס לאפיון של כל אחד מקירות המבנה – קירות המעטפת וקירות פנימיים, והחומרים מהם הם עשויים וכן לחילופי האוויר בין הדירה וסביבתה ובין חדרי הדירה לבין עצמם. הרצת ממצאי הבדיקות במודל הקרינה ההולנדי תאפשר להעריך באופן מציאותי את תרומת אפר הפחם כתוסף לבטון למנת הקרינה לה נחשף אדם מייצג מהציבור, ושל אוכלוסיות משנה, בהנחות שונות על משך השהייה בכל אחד מחדרי הדירה. בעזרת המודל ההולנדי ניתן יהיה גם להעריך באופן מושכל את החשיפה של ילדים, הרגישים לקרינה מייננת יותר מבוגרים, המשוכנים בהנחת מחדל בממ"דים.

לוט: מסגרת עבודה – חישוב מנת קרינה בדירה ישראלית
השוואה בין מעבדתית שורק – NRG
Impact from Fly Ash as Additive to Concrete on the Radiation Exposure in Dwellings, G. de With, Journal of Nuclear Engineering and Radiation Science, July 2017, Vol. 3
A comparison of methods for the determination of the natural radioactivity content and radon exhalation, G. de With et al, Radiation Measurements 105 (2017) 39e46

note

to	:	Omri Lulav	NCAB
from	:	Govert de With	NRG
date	:	6 October 2017	
reference	:	K6210/17.145365	
subject	:	Proposal for modelling the annual effective dose from external radiation and radon for a typical Israeli apartment	

Introduction

This documents provides a proposal for calculating the annual effective dose from external radiation and radon in a typical Israeli apartment. The calculation of the effective dose is based on calculation methods that consider key characteristics of the apartment, such as: floor plan, ventilation of the apartment and safe room, and selected building material. Calculations will be performed for all 21 types of concrete that have been tested by NRG[1] since 2011.

Details of the proposed modelling and a listing of all scenarios that will be calculated is provided in the section below.

Modelling approach

External radiation

The modelling approach for computing the external dose (E_{Ex}) is as follows:

- The computations will be performed using the radiation software MicroShield®. MicroShield® is a comprehensive photon/gamma ray shielding and dose assessment program that is widely used for designing shields, estimating source strength from radiation measurements and minimizing exposure to people.
- All rooms including corridor and bathroom will be represented by a rectangular cuboid with a height of 2.7m, as shown in Figure 1.

- For each room the presence of doors and windows will be geometrically accounted for in the computation.
- A 20 cm thickness will be assumed for all external walls, ceiling and floor. The inner walls will be 10 cm thick. As the 10 cm does not provide full shielding from the neighbouring rooms its effect will be estimated.
- The concrete density of the external walls is variable and based on the density of the test samples. The density of the inner walls is 1500 kg m^{-3}.
- The dose rate per hour will be determined in the centre of the cuboid.
- The annual effective dose will be determined assuming an 80% occupation time that corresponds with 7000 hr per annum for each of the cuboids.
- The effective dose rates and annual dose will be computed for all 21 concrete mixtures reported by NRG[1]. The dose contribution from fly ash will be computed by subtraction of the reference concrete from the concrete with fly-ash.

Figure 1 Schematic overview of the model boxes. The estimated boxes are rectangular with a height of 2.7m, dose points are highlighted in blue-red circles.

A summary of the calculations that will be performed for each concrete mixture is presented in the table below.

Table 1 Summary of the calculations for external dose, as performed for each of the 21 concrete mixtures.

Dose rate (µSv/h) per hour for each room.
Annual effective dose (mSv), based on an occupation time of 7000 hr per annum.
Contribution from fly-ash to the annual effective dose (mSv) for each concrete mixture with fly-ash.

Radon and radon progeny

The modelling approach for computing the radon dose (E_{Rn}) is as follows:

- A multi box model will be constructed to compute the radon and radon progeny concentrations in the apartment.
- A radon concentration for the outdoor environment will not be included.
- The number of compartments and the ventilation rate applied in the box model is as follows:
    - For a regular situation the apartment will be considered as one space, where the air exchange with outdoor is 0.5 to 1 h^{-1} during night (9 hours a day) and 1 to 3 h^{-1} during the rest of the day.
    - For an extreme situation, air exchange between 'Child bedroom & Safe room' and the rest of the apartment during night should be assumed 20 m^3 h^{-1}, and no air exchange with the outdoor environment.
- The radon source term will be based on the total surface area of the concrete, including external walls, floor and ceiling. Where the measured exhalation rate will be corrected for the presence of three-dimensional exhalation effects.
- Radon and radon progeny concentrations for the regular situation will be computed using a min, max and mean scenario. For an extreme situation during the night 20 m^3 h^{-1} will be applied to the 'Child bedroom & Safe room' combined with a minimum ventilation in the remaining of the apartment.
- The annual dose from radon for the regular situation will be based on an occupancy time of 7000 h.
- The annual radon dose for the regular and extreme situation will be computed for all 21 concrete mixtures reported by NRG[1]. The dose contribution from fly ash will be computed by subtraction of the reference concrete from the concrete with fly-ash.

A summary of the calculations that will be performed for each concrete mixture is presented in the table below.

Table 2 Summary of the calculations for radon, as performed for each of the 21 concrete mixtures.

Regular situation	
Minimum ventilation	0.50 h^{-1} (night) / 1.0 h^{-1} (day)
Mean ventilation	0.75 h^{-1} (night) / 2.0 h^{-1} (day)
Maximum ventilation	1.00 h^{-1} (night) / 3.0 h^{-1} (day)
Extreme situation	
Minimum ventilation	20 m^3 h^{-1} (night) / 1.0 h^{-1} (day)
Annual radon dose (mSv) for all above listed situations, based on an occupation time of 7000 hr per annum.	
Contribution from fly-ash to the annual radon dose (mSv) for each concrete mixture with fly-ash.	

The dose results from external radiation and radon will be combined to provide for an estimate of the total dose.

Reporting of the results

The results will be reported in a technical document, including a description of the methodology, details of the input parameters and a summary of the results with conclusions.

References

[1] De With, G. (2017)
Radiological characterization and dose assessment of Israeli concrete - Survey 2011 – 2016, NRG-report 913155/17.144627, Nuclear Research and consultancy Group, Arnhem, The Netherlands.

[2] ICRP (1994)
Human respiratory tract model for radiological protection. Publicatie 66. International Commission on Radiological Protection. Pergamon Press, Oxford.

סקר קרינה מתערובות בטון נפוצות
השוואה בין-מעבדתית

שנה	מוצר	מנת קרינה שנתית (mSv/y)*					
		רכיב גמא		רכיב רדון		כולל	
		ממ״ג-שורק	NRG**	ממ״ג-שורק	NRG	ממ״ג-שורק	NRG
2013	בטון עם אפר	0.25 - 0.2	0.27 - 0.22	0.46 - 0.42	0.68 - 0.62	0.71 - 0.62	0.96 - 0.84
	בטון ייחוס	0.15	0.19	0.39	0.63	0.55	0.82
2014	בטון עם אפר	0.28 - 0.21	0.29 - 0.23	0.42 - 0.39	0.63 - 0.62	0.7 - 0.62	0.92 - 0.86
	בטון ייחוס	0.17	0.18	0.45	0.61	0.62	0.79
2015	בטון עם אפר	0.27 - 0.21	0.26 - 0.22	0.43 - 0.31	0.63 - 0.48	0.7 - 0.52	0.88 - 0.7
	בטון ייחוס	0.15	0.18	0.41	0.57	0.56	0.76
2016	בטון עם אפר	0.28 - 0.2	0.26 - 0.22	0.46 - 0.43	0.64 - 0.63	0.74 - 0.63	0.91 - 0.85
	בטון ייחוס	0.17	0.19	0.43	0.6	0.6	0.78

* תחום של תערובות בטון עם מקורות אפר (כתוסף לבטון בשיעור 100 ק״ג/מ״ק) בדידים נבחרים ותערובות בטון ייחוס ללא אפר כתוסף.

** החישובים נעשו לפי תקן הקרינה ההולנדי ושיטות המדידה של התקן, המבוססות על נסיון רב-שנים ונבחנו במחקרים ובפרסומים רבים בעיתונות מקצועית בינלאומית. המדידות בוצעו על ידי מעבדת NRG לבטיחות קרינה, הולנד, עם למעלה מ- 40 שנות נסיון.

לעיון בממצאי הבדיקות של NRG לתערובות סקר 2012 ואילך, בקישור להלן.

Impact From Fly Ash as Additive to Concrete on the Radiation Exposure in Dwellings

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Fly ash is widely used as a supplementary cementitious material in the production of cement and concrete, and improves durability and strength of the concrete. However, as for all materials of mineral origin, fly ash is a source for natural radioactivity; hence, its need for responsible use. The aim of this study is to investigate the radiation impact from fly ash as an additive to concrete compared against concrete without fly ash. For this purpose, eight concrete mixtures are experimentally tested, followed by a computation of the radiation dose when used as bulk material in building constructions. The results demonstrate an increase in the total radiation dose from around 0.8 mSv with no fly ash up to 0.92 mSv when fly ash is used. The increase mostly comes from external radiation, while the radon exhalation factor is reduced and sometimes even reduces the radon dose despite the higher radium content. The work has demonstrated that the impact from fly ash on the radiation exposure is limited when applied as a supplementary cementitious material. At the same time, fly ash provides real benefits to the quality and durability of the concrete. For this reason, exemption strategies for such applications should be developed. [DOI: 10.1115/1.4036322]

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. The most important benefit is its reduced permeability to water and aggressive chemicals. Furthermore, properly cured concrete made with fly ash creates a denser product because of reduced pore size. This increases strength and reduces permeability. However, as for all materials of mineral origin, fly ash is a source for natural radioactivity; hence, its use in building materials is regulated [1] with numerous guidelines on its responsible use [2]. For example, the EU Basic Safety Standard also known as EU-BSS [1] now sets specific requirements on the received external dose from building materials and lists fly ash as a material to be considered for its 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 determine the radiation impact from concrete used in building construction with fly ash additive compared against concrete without. For this purpose, a total of eight different concrete mixtures are studied, including mixtures with and without 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 paper first describes the methods to determine the radiological properties of the concrete mixtures and the modeling techniques used in the assessment. The method description is followed by an overview of the results and a discussion on the outcome. The paper ends with a summary of the main findings.

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Testing and Modeling Methods

Experimental Testing. As part of the presented work, a total of eight 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. The eight concrete mixtures are tested in two sets. Each set contains one mixture without fly ash followed by three mixtures containing fly ash of different origins. The fly ash content in each mixture is broadly similar for all cases and is approximately 100 kg/m^3. An overview of the concrete mixtures is shown in Table 1. For this purpose, concrete samples were provided in dual. The activity content was determined in three identical samples ($0.1 \times 0.1 \times 0.1$ 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 is measured in a single test using a separate set of three identical samples ($0.1 \times 0.1 \times 0.2$ m^3).

Activity Concentrations. The natural radioactivity concentrations of the specimens are determined according to a standard method published under NEN 5697 [3,4]. According to this method, the density-dependent photopeak efficiencies are

Table 1 Overview of the tested samples

Sample	Fly ash (kg/m^3)	Density (kg/m^3)
1.1	—	2420
1.2	100	2420
1.3	100	2418
1.4	100	2422
2.1	—	2512
2.2	100	2518
2.3	100	2537
2.4	100	2494

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 1461 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 l, weighted, and closed radon-tight. To obtain secular equilibrium, a waiting time of at least 3 weeks is taken into account before counting the samples. All samples are counted using a high purity germanium detector in a low-background facility. The samples of the material are analyzed in an identical way as the calibration standards with respect to geometry, waiting time, and radon-tightness of the beaker. The photopeak 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.

Radon Exhalation Rate. The natural free radon exhalation rate 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 l 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 0–100% by means of a controlled mixing of dry and water–saturated nitrogen gas (Fig. 1). After a given time (normally within 3 h), a steady-state concentration is reached, and the experiment can be started. The outcoming flow is guided through two U-shaped tubes for a period of 10–30 min. 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.

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/s Bq can be reached.

The samples are conditioned at a temperature of $20 \pm 2\,^{\circ}$C and a relative humidity of $50 \pm 5\%$. Equilibrium is achieved when the mass of the sample over a period of 7 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.

Modeling Approach

Radon and Radon Progeny. In this work, a computational fluid dynamics (CFD) model is used to simulate the concentration of radon and radon progeny products in a typical Israeli room. 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 modeling technique are described by De With and De Jong [6].

Dose Modelling. 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 5×4 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: gray per hour) in a particular room is then calculated according to

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

in which i is the index for a construction part, F_1 to F_n are the correction factors for each construction part i, 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: $F_{\mathrm{dose},}$ $i = k_1 \cdot a_{1,i} + k_2 \cdot a_{2,i} + k_3 \cdot a_{3,i}$. In this equation, k_1, k_2, and k_3 represent the specific absorbed dose rates, and $a_{1,i}$, $a_{2,i}$, and $a_{3,i}$ represent the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K of construction part i (Bq kg^{-1}), respectively. The values of the specific absorbed dose rate depend among 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 per Bq/kg, respectively [10]. The absorbed dose is multiplied with a conversion factor of 0.7 Sv/Gy to obtain the effective dose.

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

$$E_{\mathrm{Rn}} = \mathrm{DCF}_{\mathrm{Rn}} \cdot \mathrm{EEC}_{\mathrm{Rn}} \cdot t_{\mathrm{Rn}} \quad (2)$$

where $\mathrm{DCF}_{\mathrm{Rn}}$ is the conversion factor of 9 nSv/h per Bq/m, $\mathrm{EEC}_{\mathrm{Rn}}$ is the equilibrium equivalent ^{222}Rn concentration, and t_{Rn} is the hours per year spend indoors. The $\mathrm{EEC}_{\mathrm{Rn}}$ is calculated as 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), respectively. These concentrations are obtained from the CFD computations.

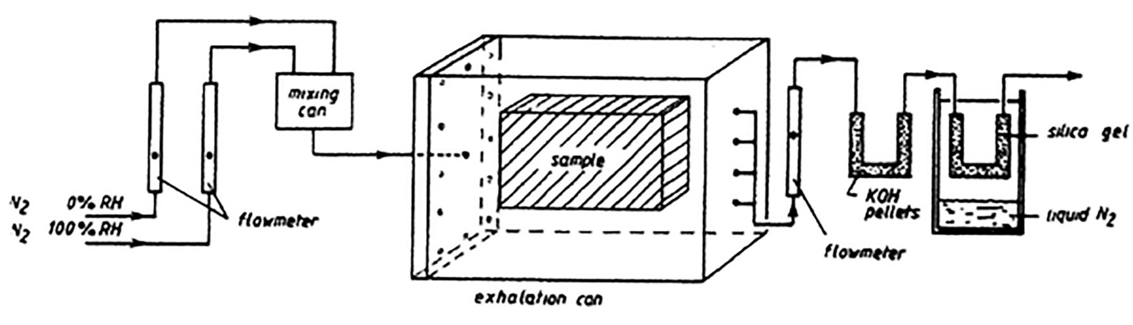

Fig. 1 Schematic view of the measuring arrangement for determination of the radon exhalation rate according to NEN 5699

Table 2 Activity concentrations with its standard uncertainty (±1 SD) expressed in Bq/kg[1] and radon exhalation rate with its standard uncertainty (±1 SD) expressed in μBq/s and μBq/(kg s)

	Activity concentrations					Radon exhalation		
						ER_{Rn}		
Sample	Density (crushed) (kg/m^3)	^{226}Ra (Bq/kg)	^{228}Ra (Bq/kg)	^{228}Th (Bq/kg)	^{40}K (Bq/kg)	(μBq/s)	(μBq/(kg s))	ER_f (%)
1.1	1600	29 ± 2	6 ± 1	5.6 ± 0.4	48 ± 3	101 ± 4	6.9 ± 0.2	11.3
1.2	1514	37 ± 2	12 ± 1	12 ± 1	65 ± 3	104 ± 2	7.2 ± 0.1	9.3
1.3	1586	35 ± 2	9 ± 1	9 ± 1	70 ± 4	106 ± 8	7.3 ± 0.5	9.9
1.4	1500	41 ± 3	17 ± 1	16 ± 1	56 ± 3	106 ± 6	7.3 ± 0.4	8.4
2.1	1656	31 ± 2	6 ± 1	5.9 ± 0.4	48 ± 3	94 ± 3	6.2 ± 0.2	9.6
2.2	1626	38 ± 3	14 ± 1	13 ± 1	65 ± 3	98 ± 11	7 ± 1	8.2
2.3	1650	37 ± 2	7 ± 1	6.4 ± 0.4	70 ± 4	69 ± 4	4.5 ± 0.2	5.8
2.4	1663	41 ± 3	10 ± 1	10 ± 1	56 ± 3	110 ± 4	7.3 ± 0.2	8.5

Results

Experimental Results. A gamma-spectrometric analysis on the radioactivity concentrations of the gamma-ray emitting radionuclides is carried out in three samples of each of the eight concrete mixtures. The results from these measurements are presented in Table 2. The results demonstrate a ^{226}Ra ranging between 29 and 41 Bq/kg 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 6–17 Bq/kg. 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 and 70 Bq/kg, and also here the activity in the mixtures with fly ash is higher.

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 2 and show a radon exhalation rate (ER_{Rn}) of approximately 4.5–7.3 μBq/(kg s). The radon exhalation from the mixtures with fly ash is in some cases higher but in one case significantly lower. This reduction occurs despite the higher ^{226}Ra concentrations in the mixtures with fly ash. Table 2 also shows the radon exhalation factor (ER_f), which represents the percentage of radon that is released from the material. 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/(kg s), $C_{Ra\text{-}226}$ is the radium concentration in Bq/kg, and λ_{Rn} is the radon decay constant. 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.

Modeling Results. Based on the experimental findings, the radiation exposure is computed using the previously described methods. 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/yr, which corresponds with 80% of the total time [11].

Radon and Radon Progeny. CFD calculations are performed for the eight concrete mixtures. The modeling 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. For this purpose, the measured radon exhalation 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 is 10 Bq/m with an equilibrium factor between radon and its progeny of 0.4 [11].

The computed concentrations for radon and its progeny are presented in Table 3. The radon concentrations are in the order of 20–30 Bq/m^3, and the progeny concentrations are reduced to around 5 Bq/m^3 for ^{214}Bi due to natural ventilation and deposition of the radon progeny. The concentrations include the radon background of 10 Bq m^{-3} and its background progeny.

Dose Modelling. Following the above described results, a dose assessment for external radiation and radon exposure is performed for the eight concrete mixtures. For this purpose, the concentrations ^{228}Ra and ^{228}Th are averaged to obtain an estimated ^{232}Th concentration in the building material. The annual effective dose from the building materials and the background radon is shown in Table 3. The results show a dose from external radiation of around 0.18–0.29 mSv/yr. The external radiation dose is higher for the samples with fly ash due to its increase in activity concentrations.

The radiation dose from radon is around 0.6 mSv/yr and includes radon background. As a result, the dose from radon is considerable higher than the dose from external radiation. However, the variation in radon dose from the different concrete mixtures is very limited. By accumulating both doses, the total dose from external radiation and radon is around 0.8 mSv with a maximum of approximately 0.9 mSv/yr for certain types of concrete with fly ash.

Discussion

The use of fly ash as an additive to concrete has resulted in an increase in the activity concentrations for all studied nuclides. These findings are consistent with earlier work reported by many researchers in the field [13]. However, the increase is moderate due to its limited use in the cement only. Based on the measured activity concentrations, the external exposure from the concrete mixtures is estimated to be in the order of 0.25 mSv/yr. This is well below the reference level of 1 mSv/yr from building materials as defined in the EU-Basic Safety Standard [1]. Even if one considers the moderate increase in external exposure from the use of fly ash, its annual dose still remains well below the 1 mSv.

The radon dose is received from only 10% of the radon exhalating from the building material. However, its resulting dose is considerably higher than from external radiation even if one excludes the contribution from the radon background. This leads to a critical note on the present regulations for natural radiation from building materials. By and large, all regulation is focused on the contribution from external radiation, while the contribution from radon is equal or even more important. If one also considers ICRP's most recent publication on radon [14], the estimated dose from radon might increase further, due to the proposed increase in the radon conversion factor. This will inevitably demand for further need to incorporate radon exposure from building materials in future regulation.

Table 3 Radon and radon progeny in Bq/m^3 and annual dose from radiation exposure in millisievert per year

Sample	Radon (progeny)				Annual effective dose		
	$C_{Rn\text{-}222}$ (Bq/m^3)	$C_{Po\text{-}218}$ (Bq/m^3)	$C_{Pb\text{-}214}$ (Bq/m^3)	$C_{Bi\text{-}214}$ (Bq/m^3)	E_{Ex} (mSv/yr)	E_{Rn} (mSv/yr)	E_{Tot} (mSv/yr)
1.1	27.9	23.7	10.1	5.2	0.18	0.61	0.79
1.2	28.4	24.2	10.2	5.3	0.25	0.62	0.87
1.3	28.7	24.4	10.3	5.3	0.23	0.63	0.86
1.4	28.9	24.5	10.4	5.3	0.29	0.63	0.92
2.1	26.0	22.2	9.5	4.9	0.18	0.57	0.76
2.2	26.7	22.8	9.7	5.0	0.26	0.58	0.84
2.3	21.6	18.4	8.0	4.2	0.22	0.48	0.70
2.4	28.9	24.6	10.4	5.3	0.25	0.63	0.88

While fly ash increases the radium content, the addition of fly ash does not lead to an equal increase in radon exposure, as shown by the reduction in the radon exhalation factor (ER_f). The reason for the reduction is the sintered fly ash grain that reduces the recoil of radon atoms from the material grains itself resulting in a reduced radon emanation. The addition of fly ash also leads to a reduced permeability of the concrete, which reduces the radon diffusion through the material pores. Both these mechanisms contribute to a reduction in the radon exhalation factor.

Based on the findings of this work, the use of fly ash results in a limited increase in radiation exposure from external radiation and radon of up to 15%, and for one concrete mixture the addition of fly ash results in a reduction of 8%. The increase by and large comes from external radiation exposure, while the increase in exposure from radon is limited or in certain cases even negligible. These findings are consistent with earlier work on the use of fly ash in concrete [13].

The EU-BSS suggests that all construction products that contain naturally occurring radioactive material (NORM) residuals like fly ash and are used in building construction should be assessed on its radioactivity content regardless of the amount of residual used. This work demonstrates that the impact from fly ash on the total radiation exposure is limited when applied as a supplementary cementitious material. At the same time, fly ash provides real benefits in terms of concrete quality and durability. For this reason, exemption strategies for these kind of applications should be developed by the authorities and (inter)-national expert committees. Recently, a first exemption strategy is presented, which uses the percentage by mass of NORM residual in the end-product as a possible measure for exemption [15]. By doing so, many NORM applications that remain far below the international reference level of 1 mSv can be exempt from further testing. Such strategy should be further developed and harmonized to allow for wider application.

Conclusions

A total of eight concrete mixtures are studied to determine the activity concentration from naturally occurring nuclides as well as its radon exhalation rate. Subsequently, the external and radon dose from these mixtures is determined.

The results demonstrate an increase in the total radiation dose from around 0.8 mSv when no fly ash is used to a maximum of up to 0.92 due to fly ash. The increase in exposure mostly comes from external radiation. Fly ash in concrete reduces the radon exhalation factor, and sometimes even reduces the exhalation rate despite its higher radium content.

This work has demonstrated that the impact from fly ash on the exposure level is limited when applied as a supplementary cementitious material. At the same time, fly ash provides real benefits in terms of concrete quality and durability. For this reason, exemption strategies for these NORM applications should be developed by authorities and (inter)national expert committees.

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Nomenclature

$a_{1\ldots3,i}$ = activity concentrations for each construction part, Bq/kg
$C_{1\ldots3}$ = radon progeny activity concentrations in the indoor environment, Bq/m^3
CFD = computational fluid dynamics
E_{Ex} = external exposure component of the effective dose, Sv/yr
E_{Rn} = radon component of the effective dose, Sv/yr
E_{Tot} = total effective dose, Sv/yr
EEC_{Rn} = radon equilibrium equivalent concentration
ER_f = radon exhalation factor
ER_{Rn} = radon exhalation rate, Bq/s or Bq/(kg s)
EU-BSS = EU Basic Safety Standard
$\dot{D}_{air}$ = absorbed dose rate in air, Gy/h
DCF_{Rn} = radon conversion factor, Sv/h per Bq/m^3
F_{adjac} = correction factor for contribution from adjacent floors and dwellings
F_{dose} = dose factor, Gy/h
F_i = correction factors for each construction part
F_{zoning} = correction factor which takes internal zoning
i = index for each construction part
$k_{1\ldots3}$ = specific absorbed dose rate for each nuclide, Gy/h per Bq/kg
t_{Rn} = hours per year spend indoors, h/yr
λ_{Rn} = radon decay constant, s^{-1}

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A comparison of methods for the determination of the natural radioactivity content and radon exhalation

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HIGHLIGHTS

- Activity concentrations and radon exhalation of concrete is measured using methods applicable in the Netherlands and Israel.
- The methods are equivalent and appropriate for testing building materials that fall under radiation protection regulation.
- The methods are on these grounds also suitable for use in harmonised standards that are presently under development.

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ABSTRACT

In this work a comparison of the activity content and radon exhalation rate of five concrete mixtures is carried out by NRG (NL) and SOREQ (IL) using the measurement standards applicable in the Netherlands and Israel, respectively. Comparison of the activity concentrations obtained by NRG and SOREQ for all concrete mixtures comply with proficiency requirements in literature and demonstrate that the results agree within a 99% confidence level. Variations in the weighted sum of the activity concentrations – computed according to the European and Israeli gamma indices – between the two laboratories are even smaller and well within the reported one standard deviation uncertainty.

The measured radon exhalation rates agree within a confidence level of 90%, despite considerable differences in the applied methods and uncertainties in the material's radon exhalation that are beyond the measurement protocol. This includes the effects from humidity and aging, and it is mentioned that future guidance on the sample representativeness would be welcomed. Based on the findings of this work it is concluded that the methods for determining the activity concentration and the radon exhalation are equivalent methods and appropriate for testing of building materials that typically fall under radiation protection regulation. Furthermore, the methods can also be recommended for use in harmonised standards that are presently under development.

1. Introduction

Fly ash – a by-product of burning pulverized coal in an electrical generating station – is widely used in the production of cement and concrete. Its potential as a supplementary cementitious material has been known almost since the start of the last century (Anon, 1914) and offers significant environmental and technological benefits in terms of reduction of carbon dioxide emission accompanying the manufacture of portland cement clinker, increased strength and reduced permeability of concrete, which improves durability of concrete structures. In this case the fly ash content of the final concrete product is usually 2–3% (by mass), assuming a 15–25% cement replacement rate (Kovler, 2012). Use of fly ash as a partial replacement of cement is favourable in structures made of mass concrete – because it reduces cement hydration heat.

As a partial replacement of sand (more accurately – of the fine fraction of sand), fly ash can be introduced in normal-weight concrete mixes by much larger amount, and this feature is especially important in the regions suffering from the lack of good quartz sand as an important concrete constituent (Kovler, 2017). In addition, fly ash as a replacement of fine sand improves workability and

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pumpability of fresh concrete mixes. At the same time, the level of 120 kg m^{-3} is usually not exceeded in concrete mixes applied in construction of habitable structures. Although high-volume fly ash (HVFA) concrete mixes containing about 200 kg m^{-3} of fly ash are known, such mixes are usually cast in pavements, foundations and road construction applications, where they have technological and economic success. However, these applications are not of our concern from a radiological point of view (Kovler, 2012).

As for all materials of mineral origin, fly ash is a source for natural radioactivity. To limit radiation exposure from building materials restrictions are imposed on the use of radioactive material from natural origin through national legislation (IM, 2017; SI, 2010). Such regulation is commonly based on international guidelines as there are the International Basic Safety Standard (BSS) (IAEA, 2014) and the EU BSS (EC, 2013). For example, the EU Basic Safety Standard sets specific requirements to limit the external dose from building materials due to its presence of ^{226}Ra (radium), ^{232}Th (thorium) and ^{4} K (potassium). These radionuclides are a source of gamma-emitting (decay) products, which will contribute to the external radiation dose. Other regulations also take account of the internal radiation dose from radon (ÖNORM, 1998; SI, 2010) due to the presence of ^{226}Ra – the parent nuclide of radon – which contributes to the radon exposure in dwellings.

Pivotal in good regulatory control are robust measurement protocols that provide for material testing with good repeatability and limited uncertainty as its results are needed to assess the radiation dose from building materials and ensure compliance. This includes measurement of the radioactivity concentration from ^{226}Ra, ^{232}Th and ^{4} K as well as the radon exhalation rate. Various national standards and protocols on the measurement of the activity content from natural radioactivity exist (SSM, 1998; UNI, 1999; NEN, 2001a; SI, 2010). Recently the EU has drafted a Technical Specification (CEN, 2017) on the measurement of ^{226}Ra, ^{232}Th and ^{4} K from building materials, and it is envisaged to be published as a European Norm (EN) by 2017. Measurement of radon exhalation rate is also subject of numerous national standards and protocols (NEN, 2001b; SI, 2010) and recently the International Standardization Organisation (ISO) has completed an ISO norm on this theme (ISO, 2016).

The purpose of this work is to perform a comparison on the measurement of the natural activity concentrations and radon exhalation rates from various concrete mixtures with and without fly ash, using the measurement standards applicable in the Netherlands and Israel, respectively. The standards in the Netherlands and Israel are well tested (Blaauw et al., 2001; Haquin et al., 2010; Kovler, 2011) and published by their national standardization organisations. As a result, the standards have a good international standing, e.g. the Dutch standards on natural radioactivity formed the basis for the international EN (CEN, 2017) and ISO (ISO, 2016) standard. For this reason, the proposed comparison will give insight in the consistency of the measured radiation properties that can be expected when assessing regular building materials for the purpose of regulatory control.

In this study a total of five types of concrete are prepared and analysed by the two different laboratories. The laboratories performed measurements according to their own national methods and protocols. Subsequently, the results are compared and a proficiency test is performed to investigate consistency in the measurement data reported by the laboratories.

2. Materials and methods

A series of measurements are performed to determine the activity concentrations of ^{226}Ra, ^{232}Th (^{228}Ra, ^{228}Th) and ^{4} K, and radon exhalation rate from five different concrete mixtures. The measurements are performed by the two nuclear research institutes, NRG and SOREQ. The measurements by NRG are performed in accordance with the Dutch NEN standards NEN-5697 (NEN, 2001a) and NEN-5699 (NEN, 2001b) for activity content and radon exhalation. SOREQ has performed its measurements according to the Israeli standard SI-5098 (SI, 2010).

2.1. Samples

The set of concrete mixtures consists of a reference mixture without fly ash followed by four mixtures containing fly ashes of different origin. For this purpose two sets of samples are prepared. Each laboratory received a set and measured the activity content in three identical samples (0.1 × 0.1 × 0.1 m^3) per concrete mixture that were crushed by the laboratory. The radon exhalation rate is measured in a single test using a separate set of three identical samples (0.1 × 0.1 × 0.2 m^3). These samples were circulated between the laboratories to perform the radon measurements on the same sample material. The fly ash content in each mixture is broadly similar for all cases and is approximately 120 kg m^{-3}, which corresponds with around 40% of the cement content. An overview of the concrete mixtures together with the origin of the fly ash is shown in Table 1. In addition a total of two concrete samples (A.1 and A.2) have been tested on their radon exhalation rate twice to demonstrate the effects from aging. The samples have also dimensions of 0.1 × 0.1 × 0.2 m^3 and are tested after respectively 6 and 23 months.

2.2. Determination of the activity concentrations

2.2.1. NEN 5697

The natural radioactivity concentrations of the specimens are determined according to a standard method published under NEN 5697 (NEN, 2001a). 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 1461 keV (^{4} 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 ^{4} K. The standards are placed into Marinelli beakers with a volume of about 1 L, 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. Prior to the measurements,

Table 1
Overview of the tested samples.

Sample	Density ($kg \cdot m^{-3}$)	Fly ash ($kg \cdot m^{-3}$)	Origin (–)
1	2420	–	–
2	2440	120	Indonesia
3	2420	120	Australia
4	2430	120	Russia
5	2430	120	Colombia
A.1	2370	–	–
A.2	2260	140	South Africa

the samples are crushed to pieces with a particle size smaller than 1 mm.

The standard method includes a test for the determination of the tightness of the sealed Marinelli beaker. In this test about 500 Bq or more of ^{222}Rn gas is injected into an almost closed beaker using a gas syringe, where after the beaker is sealed in the usual way. The beaker is counted for at least ten successive time periods of 4 h. From the time-dependence of the count rate at the 609 keV photon peak of ^{214}Bi, the leakage rate is calculated. This factor should satisfy the following inequality:

$$\lambda_L + 2s_A < 0.1\lambda_{Rn}. \tag{1}$$

where λ_L is the ^{222}Rn leakage rate, s_A the Standard Deviation (SD) of this factor as determined by the method of least squares and λ_{Rn} the decay constant of ^{222}Rn ($2.1 \cdot 10^{-6}$ s^{-1}). In case the beaker leaks at its maximum allowed rate of 0.1 λ_{Rn} the ^{222}Rn concentration in the beaker will deviate from its equilibrium concentration by a factor of $1/(1 + 0.1) = 0.91$ or 9%. If the assumed emanation factor of the building material is less than 50%, the potential under estimate in the ^{226}Ra activity concentration of the sample will be smaller than 50% of that 9% or no more than about 5%.

Prior to publication, the standard method was tested in an interlaboratory exercise. The results of that study are published by Blaauw et al. (2000, 2001).

2.2.2. SI 5098

The activity concentrations of radionuclides from natural origin were also determined according to the Israel Standard SI 5098 (SI, 2010). According to this standard the activity concentrations of ^{226}Ra, ^{232}Th and ^{4} K are determined by gamma spectrometry. The density dependent photo peak count rates of two short lived radon decay products (RDP) ^{214}Pb and ^{214}Bi (i.e. 295, 352, 609, 1,120, 1764 keV) are weighted averaged for the quantification of ^{226}Ra. The decay products in the ^{232} Th day chain are normally found in secular equilibrium in materials from terrestrial origin. Its quantification is achieved by averaging ^{208}Tl, ^{212}Pb and ^{228}Ac (photo peaks i.e. 238, 583, 911 and 2615 keV) activities concentrations. ^{4} K is quantified by its only gamma ray (1461 keV). The gamma spectrometry detector is calibrated using a multiline gamma standard source of identical geometry as the sample to be measured. The sample container is an air-tight cylindrical beaker. Prior to the measurements, the samples are crushed with a particle size smaller than 1.18 mm, homogenised, weighed and closed radon-tight to achieve secular equilibrium of the RDP's after waiting for at least three weeks. A test for the determination of the tightness of the sealed container is performed using a sample of certified uranium ore (IAEA RGU) at secular equilibrium with ^{226}Ra with an activity of about 1300 Bq. The beaker is enclosed in a previously flushed ^{222}Rn-free hermetically sealed cell of ca. 30 L where the ^{222}Rn is monitored during at least seven days. From the ingrowth curve, the maximum ^{222}Rn concentration in the cell is calculated. The leakage rate from the sample beaker is then calculated. The maximum leakage rate allowed is up to 5% (i.e. achieving 95% of secular equilibrium in the air-tight sample container).

Density correction factors are calculated when the standard source and the sample have not the same density and elemental composition. At SOREQ the correction factors as well as true coincidence summing factors are calculated using the Monte Carlo based GESPECOR software (CID Media GmbH).

2.3. Determination of the radon exhalation rate

2.3.1. NEN 5699

The natural free radon exhalation rate of the concrete samples is determined according to the standard method published under NEN 5699 (NEN, 2001b). Determination of the radon exhalation rate is based on the 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 L 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 was regulated within the full range of 0–100% by means of a controlled mixing of dry and water–saturated nitrogen gas. After a given time (normally within 3 h) a steady-state concentration in the chamber is reached and the experiment can be started. The out coming flow is guided through two U-shaped tubes for a period of 10–30 min. 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 (Darall et al., 1973).

Radioactive equilibrium in the decay chain is attained after around 3 h, and a further 13 h is required before the radon is fully released from the silica gel and diffused into the scintillation liquid. Subsequently, a recording of the energy spectrum is performed using a liquid scintillation spectrometer. For an optimal count rate the window settings should be set from 110 to 600 keV. Under ideal conditions a counting efficiency of approximately 2.8 $c \cdot s^{-1} \cdot Bq^{-1}$ can be reached. The background count rate under these conditions is around 0.15 $c \cdot s^{-1}$.

The standard conditions for the determination of exhalation rate were selected to be: 600 mL min^{-1} nitrogen flow rate, 50% relative humidity and a temperature of 20 °C, with a waiting time of 2–3 h before sampling with an absorption time of 10–30 min for the silica gel. A waiting time of 16 h and a counting time of 1 h for the LSC vials are recommended.

The method describes good repeatability and reproducibility as determined by a round-robin test (De Jong et al., 2005). In addition, the low limit of detection of 11 mBq ^{222}Rn offers the opportunity to quantify the exhalation rate of almost all kinds of mineral-based building materials. Furthermore, the advantages of this method is that it determines the free area exhalation rate and the radon detection is not influenced by external factors such as humidity.

Prior to the analyses, 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.

2.3.2. SI 5098

The radon exhalation rate of the concrete samples is calculated from the measured emanation coefficient according to the standard method published under SI 5098 (SI, 2010). According to SI 5098, this is a two-fold method; first determine the average activity of radon created by averaging the results from three identical specimens and then measure the radon activity released from the samples. The quotient of these two measures determines the emanation coefficient. The radon created in the sample is calculated from the ^{226}Ra activity concentration multiplied by the dry weight of the sample.

The radon released from the sample is measured using $0.1 \times 0.1 \times 0.2$ m^3 concrete samples by the closed chamber method. Prior to the measurement the sample is pre-conditioned to achieve equilibrium in terms of temperature and relative humidity between

the sample and the laboratory environment and sealed from all sides surfaces in the long axis with a previously proven sealing material. The specimen is placed in a hermetically closed chamber for few days with a radon detector. The chamber must have a proven sealing capability better than one air-exchange every 1500 h. At SOREQ laboratory the radon detector used consist of a pulse-height ionization chamber model Alphaguard PRO 2000 (Saphymo Ltd.) used in diffusion mode. The detector records the radon concentration in the chamber (~70 L) for at least five consecutive days. Subsequently, a time dependent accumulation of radon in the sealed chamber *C(t)* ($Bq \cdot m^{-3}$) is constructed using an ingrowth curve of the type:

$$C(t) = C_{lab}(0) \cdot e^{-\lambda_{eff} \cdot t} + C_{\max} \cdot \left(1 - e^{-\lambda_{eff} \cdot t}\right) \quad (2)$$

where C_{lab} (0) is the laboratory radon concentration ($Bq \cdot m^{-3}$) at the beginning of the measurement, λ_{eff} (s^{-1}) is the effective radon decay composed by the ventilation rate and the radon decay constant $\lambda_{eff} = \lambda_v + \lambda_{Rn}$, $C_{\max}$ is the saturated radon concentration in the chamber.

A specimen measured in a chamber of volume V (m^3) with a mass m (kg) having a homogenous constant radon concentration at the boundary (neglecting back-diffusion), the free exhalation rate, E_{Rn} ($Bq \cdot kg^{-1} \cdot s^{-1}$), is given by:

$$E_{Rn} = \frac{C_{\max} \cdot \lambda_{eff} \cdot V}{m} \quad (3)$$

A non-linear regression of the continuously monitored data according to equation (3) yields the values of $C_{lab}(0)$, $C_{\max}$, and λ_{eff}.

2.4. Data processing

For comparison of the above described test methods the experimental findings together with its uncertainty will be compared. In addition the results will be further processed for a more extensive comparison using the below presented statistical and physical parameters.

2.5. Proficiency parameter

To determine if the experimental findings from the two laboratories are statistically different a proficiency parameter u_{test} (−) is applied to the test results according to the formula by Brookes et al. (1985):

$$u_{test} = \frac{|C_{NRG} - C_{SOREQ}|}{\sqrt{u_{NRG}^2 + u_{SOREQ}^2}} \quad (4)$$

where C is the activity concentration ($Bq \cdot kg^{-1}$) and u the total uncertainty ($Bq \cdot kg^{-1}$). According to.

Brookes et al. (1985) the results agree within a 99% probability level when $u_{test} < 2.58$. For values higher than 2.58 the results are significantly different.

2.6. Gamma indices

To compare the weighted sum of the experimentally obtained activity concentrations, two gamma indices will be applied. The index according to the EU-BSS is defined as:

$$I_{BSS} = C_{Ra\text{-}226}/200 + C_{Th\text{-}232}/200 + C_{K\text{-}40}/3000, \quad (5)$$

And assumes a single building material with a surface thickness of 0.2 m and a material density of 2350 kg m^{-3}, which corresponds with a surface density of 470 kg m^{-2}. In contrast the Israeli index is a function of the materials surface density; when a surface density of 485 kg m^{-2} is selected the index reads as:

$$I_{SI} = C_{Ra\text{-}226}/411 + C_{Th\text{-}232}/290 + C_{K\text{-}40}/4036. \quad (6)$$

The activity concentration C is defined in terms of $Bq \cdot kg^{-1}$.

2.7. Radon exhalation factor

The radon exhalation factor − also sometimes named emanation coefficient − represents the percentage of radon that is released from the material. The factor is computed as:

$$E_f = E_{Rn}/(C_{Ra\text{-}226} \cdot \lambda_{Rn}), \quad (7)$$

where E_{Rn} is the exhalation rate in $Bq \cdot kg^{-1} \cdot s^{-1}$, $C_{Ra\text{-}226}$ the radium concentration in $Bq \cdot kg^{-1}$ and λ_{Rn} the radon decay constant (s^{-1}).

3. Results

3.1. Activity concentrations

Radioactivity concentrations of the gamma-ray emitting radionuclides are measured in three samples of each of the five concrete mixtures. The results from these measurements are presented in Table 2. The table also includes the average concentrations for each mixture together with its standard uncertainty. The uncertainty in the mean concentration is either based on the SD of the individual results or the combined uncertainty of the individual SD's. It is the highest of the two that is reported in the table.

The results demonstrate by and large increased activity concentrations for the concrete mixtures with fly ash compared against those without. The increases are moderate particularly for ^{226}Ra with an increase of up to 30% depending on the origin of the fly ash. For ^{232}Th the increase from fly ash in relative terms is higher. Contrary to expectation, the results from SOREQ on mixture 5 demonstrate consistently lower ^{226}Ra concentration for all three samples when compared against the reference mixture. In addition, also NRG results demonstrate some unexpected variations in the activity concentration of concrete mixture 5. The concentrations in sample 5-C for all three nuclides (^{226}Ra, ^{232}Th and ^{4} K) are significantly higher than those found in sample 5-A and 5-B. It is feasible that the deviation for this specific sample (5-C) stems from issues connected with the sample preparation, such as sampling at the production site.

A comparison of the experimental findings from NRG and SOREQ is presented in three scatterplots shown in Fig. 1. The figure includes pictures with the individual results from all three nuclides. The scatter plots indicate consistency and only limited bias in the results. By and large the variations appear randomly both as an over prediction of one method against the other and vice versa. Based on a comparison of the mean activity concentrations u_{test} does not exceed a value of 2.5 with many of the concentrations reporting a u_{test} of less than 1 (Table 2). Consequently, the activity concentrations determined by NRG and SOREQ are, based on a 99% confidence level, not significantly different.

From a radiation protection perspective, it is the weighted sum of the concentrations that determines the degree of exposure when applied as a bulk material in the building construction. For this reason, the gamma indices I reported in the EU-BSS (EC, 2013) and the SI-5098 (SI, 2010) and formulated in equations (5) and (6) are applied and presented in Table 3.

According to Table 3 the index values I_{BSS} are in the range of

Table 2
Activity concentrations with its standard uncertainty (±1 SD) expressed in $Bq \cdot kg^{-1}$ and the proficiency parameter.

Sample	NRG (NEN-5697)			SOREQ (SI-5098)			Proficiency parameter (u_{test})		
	$C_{Ra\text{-}226}$ ($Bq \cdot kg^{-1}$)	$C_{Th\text{-}232}$ ($Bq \cdot kg^{-1}$)	$C_{K\text{-}40}$ ($Bq \cdot kg^{-1}$)	$C_{Ra\text{-}226}$ ($Bq \cdot kg^{-1}$)	$C_{Th\text{-}232}$ ($Bq \cdot kg^{-1}$)	$C_{K\text{-}40}$ ($Bq \cdot kg^{-1}$)	$u_{t,Ra\text{-}226}$ (−)	$u_{t,Th\text{-}232}$ (−)	$u_{t,K\text{-}40}$ (−)
1-A	34 ± 2	6.1 ± 0.3	48 ± 3	37 ± 3	5.7 ± 0.3	43 ± 4			
1-B	35 ± 2	6.6 ± 0.3	49 ± 3	38 ± 3	5.3 ± 0.4	46 ± 7			
1-C	35 ± 4	5.8 ± 0.4	46 ± 2	36 ± 2	5.2 ± 0.4	41 ± 6			
1	35 ± 2	6.2 ± 0.3	48 ± 2	37 ± 2	5.4 ± 0.2	43 ± 3	1.0	1.9	1.2
2-A	36 ± 2	8.6 ± 0.4	75 ± 3	37 ± 3	7.5 ± 0.4	64 ± 6			
2-B	33 ± 3	8.2 ± 0.6	68 ± 5	38 ± 3	7.6 ± 0.5	68 ± 7			
2-C	37 ± 2	9.0 ± 0.4	78 ± 3	38 ± 3	7.4 ± 0.5	64 ± 8			
2	35 ± 2	8.6 ± 0.3	74 ± 4	38 ± 2	7.5 ± 0.3	65 ± 4	1.0	2.5	1.4
3-A	36 ± 4	10.1 ± 0.6	48 ± 2	40 ± 3	8.9 ± 0.4	47 ± 6			
3-B	37 ± 4	10.5 ± 0.6	50 ± 2	40 ± 3	9.6 ± 0.6	49 ± 4			
3-C	37 ± 2	10.4 ± 0.4	49 ± 3	39 ± 3	9.3 ± 0.5	49 ± 6			
3	37 ± 2	10.3 ± 0.3	49 ± 1	40 ± 2	9.3 ± 0.3	48 ± 3	1.1	2.4	0.2
4-A	41 ± 2	9.8 ± 0.4	74 ± 3	43 ± 3	8.4 ± 0.4	66 ± 5			
4-B	40 ± 2	8.6 ± 0.4	73 ± 3	42 ± 3	7.8 ± 0.5	67 ± 7			
4-C	40 ± 2	9.4 ± 0.6	73 ± 3	41 ± 3	9.0 ± 0.5	69 ± 7			
4	40 ± 1	9.2 ± 0.5	73 ± 2	42 ± 2	8.4 ± 0.5	67 ± 4	0.8	1.2	1.5
5-A	36 ± 2	9.3 ± 0.4	55 ± 3	34 ± 3	11.3 ± 0.7	62 ± 9			
5-B	38 ± 2	8.8 ± 0.6	59 ± 3	35 ± 3	10.4 ± 0.7	68 ± 10			
5-C	47 ± 4	11.1 ± 0.6	72 ± 3	34 ± 2	8.5 ± 0.4	54 ± 4			
5	40 ± 5	10 ± 1	62 ± 7	34 ± 2	10 ± 1	61 ± 6	1.2	0.2	0.1

0.16–0.20, while the index values I_{SI} are lower and range from 0.12 to 0.15. More importantly, the index values are broadly similar for NRG and SOREQ, with minimal deviations. A scatterplot of the findings is presented in Fig. 2 and demonstrates that the findings from NRG and SOREQ are within one SD uncertainty. The consistency in the index values is obtained for samples with an index value of less than 0.2, which lies well below the international reference level of one mSv for building materials.

The consistency in the results obtained by both laboratories is at least in part due to the use of mature test methods. Both methods consider appropriate energy peaks to determine the activity concentrations of the radionuclide (chains) and include corrections for density to take account of self-attenuation in the sample. The radon-tightness of the beaker is tested prior to the measurement with a maximum permitted leakage of 5% under operational conditions, and a waiting time of three weeks is required to ensure equilibrium in the radium decay chain. Furthermore, crushing of the sample material is required to a particle size smaller than ~1 mm to ensure uniform distribution of the radium progeny in the beaker. As a result, all key aspects of the testing are addressed in both methods. Nevertheless, there are considerable differences in the implementation of the test procedures. For example, the density correction in the Dutch NEN standard is based on experimental testing, while the Israeli standard allow the laboratory to use its own developed and validated method. SOREQ has chosen its corrections based on Monte Carlo type simulations. Further differences are found in the assessment of the beaker's radon-tightness. The NEN standard proposes a radon-tightness test where a radon puff is released in an empty beaker. The beaker is then measured using gamma-spectrometry and the decay in radon progeny is a measure of radon-tightness. The test is performed under worst-case conditions as all radon is air borne. In contrast, the Israeli standard proposes a certified sample with ^{226}Ra to be placed in the beaker. Subsequently the release of radon from the beaker is measured in a larger cell. The measured radon concentration in the outer cell provides a measure of the radon-tightness. Based on the reported findings it is concluded that the test methods by NEN and SI — despite the above-mentioned differences in the test procedures — are equivalent and appropriate for the kind of building materials tested here.

3.2. Radon exhalation rate

The ^{222}Rn exhalation rate from five different concrete mixtures is determined according to equation (3) and the tested samples have been circulated between both laboratories to ensure measurement on identical samples. The results from the experiments are presented in Table 4 and show the radon exhalation rate (E_{Rn}) expressed in $\mu Bq \cdot s^{-1}$ and $\mu Bq \cdot kg^{-1} \cdot s^{-1}$. In addition, the table shows the radon exhalation factor (E_f) named as radon emanation by SI 5098. The highest exhalation rate is found in concrete mixture 3, while the reference concrete without fly ash has a medium exhalation rate. For the concrete mixtures 2, 4 and 5 with fly ash the measured exhalation rate is in some case even lower than the reference mixture, despite its higher ^{226}Ra concentration. However, the reduction is not always confirmed by both labs and is sometimes statistically insignificant. The exhalation factor, determined according to equation (7) is estimated to be around 6–10% for all samples. The lowest factors are found for the two concrete mixtures with fly ash (4 and 5). This is consistent with other studies that demonstrate a reduction in the radon emanation due to the addition of fly ash (Roelofs and Scholten, 1994). However, this is not the case for the other mixtures with fly ash where an increase in the exhalation factor is found. This finding is most prominent in mixture 3. A comparison of the results from NRG and SOREQ is presented in Fig. 3 and shows that the results from both labs show good comparison. The only sample that shows considerable difference is concrete mixture 5, which shows a variation of around 15–20%; however, even then the variation does not exceed two SD's uncertainty. Computation of the proficiency parameter u_{test} according to equation (4), but now adjusted for E_{Rn}, demonstrates values of up to 1.41 (Table 4) and are well below the acceptance

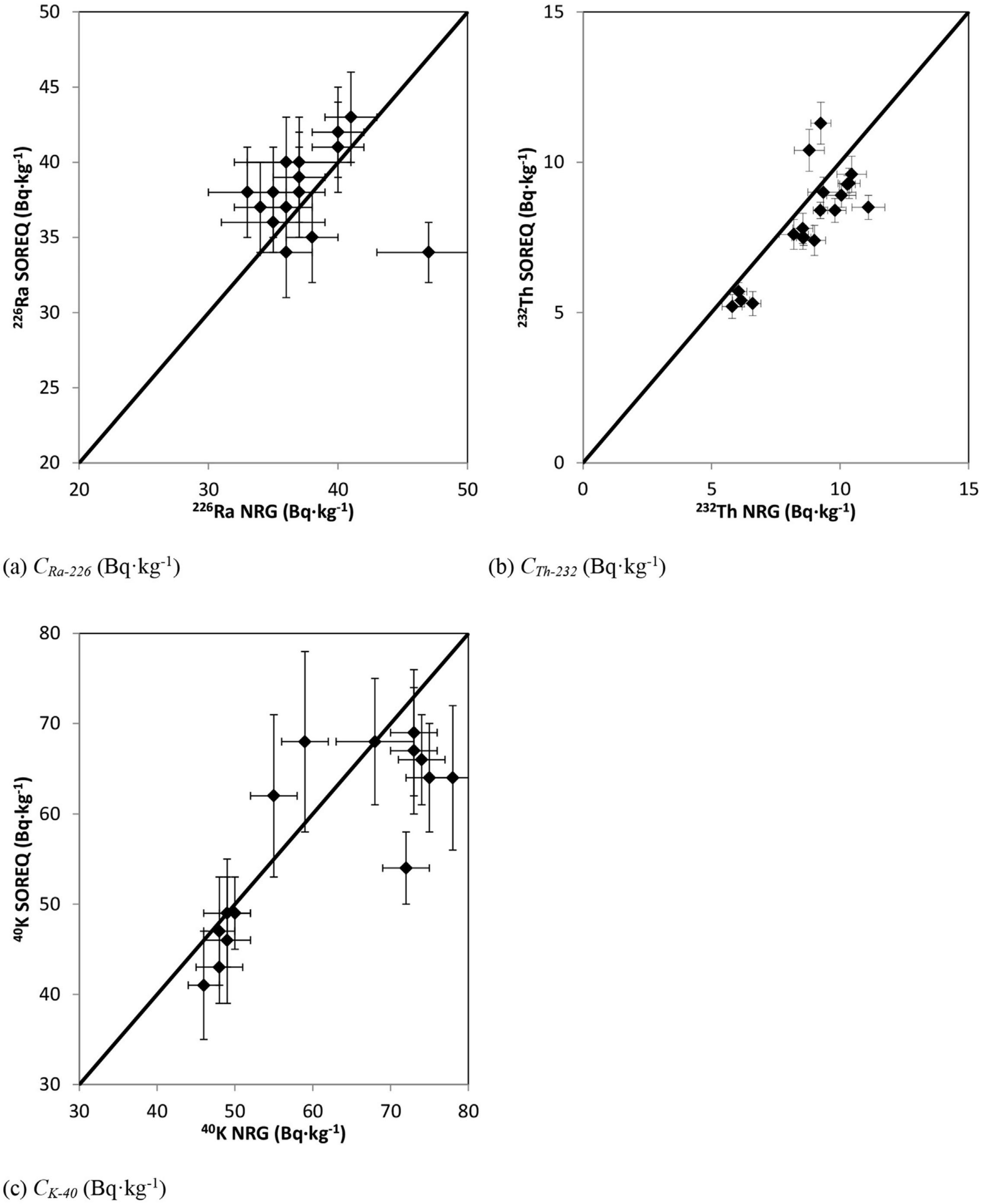

Fig. 1. Scatter plot of the NRG and SOREQ results on the activity concentrations with its standard uncertainty (±1 SD) in Bq·kg^{-1}.

Table 3
Gamma index *I* with its standard uncertainty (±1 SD) according to the EU-BSS index and the SI-5098 index.

Sample	I_{BSS} (EU-BSS)		I_{SI} (SI-5098)	
	NRG	SOREQ	NRG	SOREQ
	(−)	(−)	(−)	(−)
1	0.16 ± 0.01	0.16 ± 0.01	0.12 ± 0.01	0.12 ± 0.01
2	0.19 ± 0.01	0.18 ± 0.01	0.13 ± 0.01	0.13 ± 0.01
3	0.19 ± 0.01	0.19 ± 0.01	0.14 ± 0.01	0.14 ± 0.01
4	0.21 ± 0.01	0.20 ± 0.01	0.15 ± 0.01	0.15 ± 0.01
5	0.20 ± 0.02	0.19 ± 0.01	0.15 ± 0.01	0.13 ± 0.01

criteria of $u_{test} < 2.58$. It can be concluded that the u_{test} for each tested mixture is even below the value of 1.64 suggesting the results are within the 90% confidence level and therefore do not differ significantly.

Such findings are quite an achievement; contrary to the methods used for determining the activity content, the test methods from NRG and SOREQ for radon exhalation are fundamentally different. The method by NRG uses a purge and trap method where the radon is exhaled from the sample in an exhalation chamber that operates at low background due to its continuous ventilation with radon-free nitrogen. The radon is then

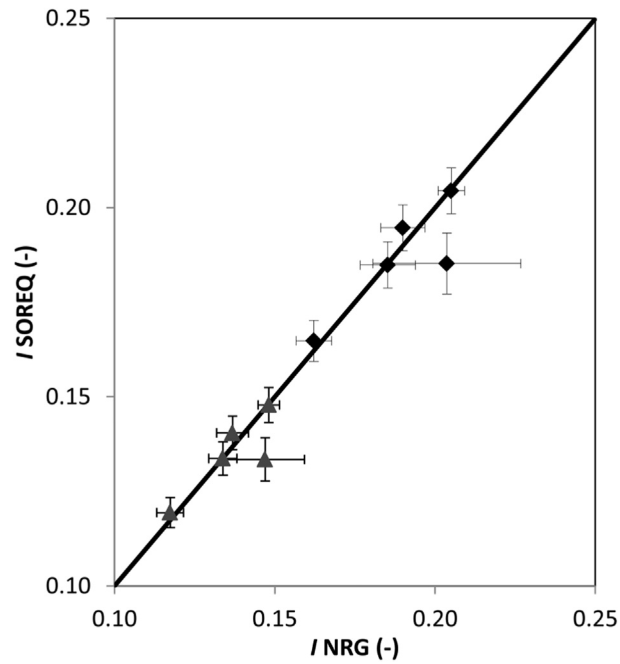

Fig. 2. Scatter plot of the NRG and SOREQ results on the gamma index with its standard uncertainty (±1 SD) in. ⬩ I_{BSS} (EU-BSS) and ▴ I_{SI} (SI-5098).

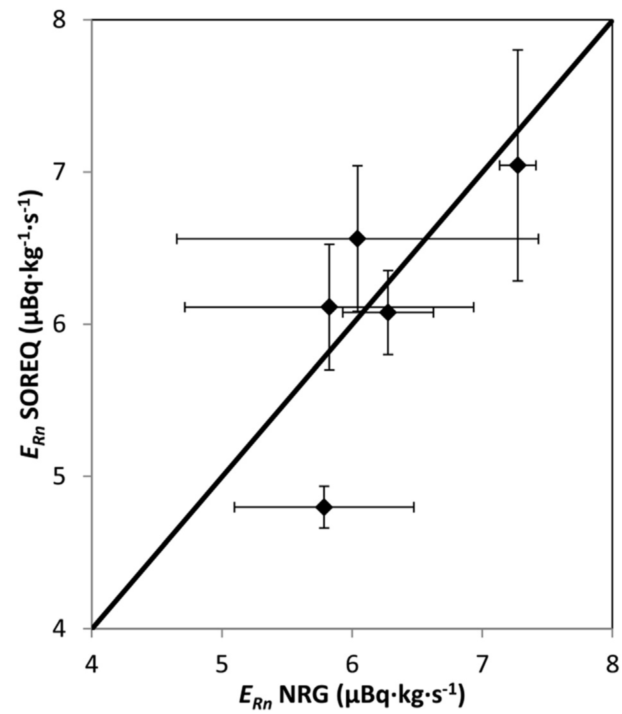

Fig. 3. Scatter plot of the NRG and SOREQ results on the radon exhalation rate with its standard uncertainty (±1 SD) in $\mu Bq \cdot kg^{-1} \cdot s^{-1}$.

captured and measured using LSC detection technique. In contrast, the method by SOREQ is based on an accumulation technique where an active radon detector measures the radon concentration in the accumulation chamber itself. Both techniques have their own challenges; the purge and trap method requires that all radon is trapped by the silica gel and that it is subsequently diffused in the scintillation liquid. The accumulation technique on the other hand requires a perfect seal of the chamber. Furthermore, as the radon concentration in the accumulation chamber increases the radon exhalation rate reduces and may lead to back diffusion. In addition, there are common challenges for both methods, which involve good temperature and humidity control of the samples. Subject to the material composition radon exhalation can be influenced considerably by these environmental conditions. Last but not least the micro structure of the concrete is influenced by hydration and carbonization. Considering that the samples are shipped across long distances and subsequently re-conditioned to the appropriate temperature and humidity, these mechanical processes form a real source of uncertainty. Despite this, there is generally good agreement between the methods.

The impact from humidity and aging on the radon exhalation from concrete has been studied previously. Earlier results from radon exhalation measurements on concrete mixtures similar to those studied in this work have demonstrated that the radon exhalation from concrete can change by as much as 35% during its first two years of age (Table 5). However, the magnitude of change as well as a possible increase or decline in radon exhalation during the first two years is dependent on many factors. These include a.o. the initial water content of the content mixture, concrete composition and humidity conditions during aging (Roelofs and Scholten, 1994). These will affect the capillary structure during its lifetime, which subsequently affects the release of radon. From a metrology perspective, this raises the question if these phenomena should be accounted for in the test protocol to allow for consistency in the measurement results or if they are an intrinsic aspect of the material design. The present NEN and IS standard addresses the issue to a limited extent by prescribing a minimum curing time before testing can start. However, this does not account for the long term effects that take place over a prolonged time even exceeding multiple years. Further work that contributes to better understanding of the effects from humidity and aging on the radon exhalation would be welcomed and should contribute to better guidance on representativeness of the test results.

Table 4
Radon exhalation rate expressed in $\mu Bq \cdot s^{-1}$ and $\mu Bq \cdot kg^{-1} \cdot s^{-1}$ and radon exhalation factor in % with its standard uncertainty (±1 SD) and the proficiency parameter.

Sample	NRG (NEN-5699)			SOREQ (SI-5098)			Pro. Para. (u_{test})
	E_{Rn}		E_f	E_{Rn}		E_f	$u_{t,E\text{-}Rn}$
	($\mu Bq \cdot s^{-1}$)	($\mu Bq \cdot kg^{-1} \cdot s^{-1}$)	(%)	($\mu Bq \cdot s^{-1}$)	($\mu Bq \cdot kg^{-1} \cdot s^{-1}$)	(%)	(−)
1	90 ± 5	6.3 ± 0.3	8.6 ± 0.6	88 ± 4	6.1 ± 0.3	7.8 ± 0.5	0.45
2	87 ± 20	6 ± 1	8 ± 2	96 ± 7	6.6 ± 0.4	8 ± 1	0.35
3	104 ± 2	7.2 ± 0.1	9.5 ± 0.5	102 ± 11	7.1 ± 0.8	8 ± 1	0.30
4	84 ± 16	6 ± 1	7 ± 1	89 ± 6	6.1 ± 0.4	7 ± 1	0.24
5	84 ± 10	6 ± 1	7 ± 1	70 ± 2	4.8 ± 0.1	6.7 ± 0.4	1.41

Table 5
Radon exhalation rate with its standard uncertainty (±1 SD) expressed in $\mu Bq \cdot s^{-1}$ and $\mu Bq \cdot kg^{-1} \cdot s^{-1}$.

Sample	Age 6 months		Age 23 months	
	E_{Rn}		E_{Rn}	
	$(\mu Bq \cdot s^{-1})$	$(\mu Bq \cdot kg^{-1} \cdot s^{-1})$	$(\mu Bq \cdot s^{-1})$	$(\mu Bq \cdot kg^{-1} \cdot s^{-1})$
A.1	149 ± 3	10.5 ± 0.2	97 ± 3	6.8 ± 0.2
A.2	74 ± 6	5.4 ± 0.5	65 ± 3	4.8 ± 0.2

4. Conclusions

The results from this work demonstrate good consistency when comparing the measured results from NRG and SOREQ. The variations in activity content are in some cases beyond the 1 SD uncertainty when considering the results from the individual radionuclides. There are some significant variations in the results from concrete mixture 5 and it is expected that these stem from issues related to the test samples. However, based on a comparison of the mean activity concentrations the acceptance criteria of $u_{test} <$ 2.58 are satisfied for each concrete mixture. Consequently, the mean activity concentrations determined by NRG and SOREQ do not differ significantly based on a 99% confidence level. When considering the weighted sum of the activity concentrations the results from the laboratories are nearly all within the reported one SD uncertainty. This is an important finding as the weighted sum is essential for the determination of the dose and compliance with legislation.

The radon exhalation rates reported by NRG and SOREQ are in close agreement and are all within a confidence level of 90% ($u_{test} < 1.64$). Most significant variations are found with mixture 5 demonstrating a variation in the measured exhalation of only 15–20%. Comparable results are found despite the use of very different test methods. The method by NRG is based on a ventilated exhalation chamber with purge-and-trap approach, while the SOREQ method is based on a direct radon measurement in a non-ventilated exhalation chamber. Furthermore, the radon exhalation is subject to additional uncertainties that are beyond the measurement protocol. Despite this the findings are consistent.

Based on the findings of this work it is concluded that the methods for determining the activity concentration and the radon exhalation are equivalent methods and appropriate for testing of building materials that typically fall under radiation protection regulation. Furthermore, The methods and their procedures are on these grounds also recommended for use in harmonised standards that are presently under development. However, some challenges particularly related to the measurement of radon exhalation do remain. These include the effects from aging and humidity. Auxiliary results presented in this work have shown the variations in radon exhalation that can occur in concrete due to aging. Such issues are beyond the measurement protocol itself, but guidance on how to deal with these phenomena is important to enable consistency in the measurement results.

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