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Research paper

Evaluation of the I-index by use of a portable hand-held spectrometer and laboratory methods - a risk assessment of Swedish concrete by use of different crushed aggregates

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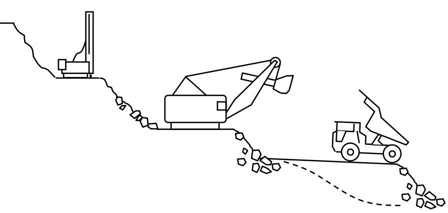
ABSTRACT

The recommended levels of ionizing radiation from construction materials in effective dose is set to a maximum of 1 mSv/year, EC (1996, 1999, 2013), ICRP (2007), IAEA (2011). By using a theoretical model proposed by the European Union (1999), this is equivalent to I-index 1. By using of concrete slabs with dimensions of 1.5 m × 1.5 m × 0.15 m, an empirical approach is suggested for the calculation of the I-index of naturally occurring ionizing radiation from construction materials. Measurements of ^{40}K, ^{226}Ra, ^{232}Th and the total gamma radiation were conducted and the I-index values were calculated for each concrete mix. A good linear relationship could be established between measurements performed by the Swedish Cement and Research Institute (CBI) and the laboratory results acquired from the Radiation and Nuclear Safety Authority of Finland (STUK) and Centre de Recherches Pétrographique et Géochimiques/ Le Centre National de la Recherche Scientifique (CRPG/CNRS). The results indicate that 60 % of the investigated construction materials are in agreement with the stipulated levels set out by the EC (1999, 2013). The cause for the higher levels of ionizing radiation is often elevated concentrations of all the radioactive elements measured. Regarding the concrete samples yielding values of I-index > 1, ^{232}Th makes the largest contribution.



1. INTRODUCTION

All construction materials contain various amounts of naturally occurring ionizing radiation. The naturally occurring radioactive nuclides used for evaluation of radiological risk assessment are Thorium-232 (^{232}Th), Radium-226 (^{226}Ra) and Potassium-40 (^{40}K), as well as the total gamma radiation within the energy range 40-2810 keV (IAEA, 2003). It is of importance for authorities and regulators, as well as for habitants, to evaluate the effects of these radioactive nuclides impact on human health and to assess the risks for use of the products aimed for dwellings (IAEA, 2011).

The European Commission (1996, 1999, 2013) has during the course of the last 15-20 years imposed clarification of the acceptable levels of naturally occurring ionizing radiation within construction materials for member states. The recommended levels from construction materials in effective dose is set to a maximum of 1 mSv/year, EC (2013), ICRP (2007), IAEA (2011). Using a model described in RP 112 (EC, 1999), the effective dose from building materials can be expressed in the form of an activity index I, where the index is calculated from the activity concentrations of ^{40}K, ^{226}Ra and ^{232}Th as follows:

I = $\mathbf{C}^{40}$K/3000 Bqkg^{-1} + $\mathbf{C}^{226}$Ra Bqkg^{-1}/300+$\mathbf{C}^{232}$Th/200 Bqkg^{-}1, where C is the activity concentration in Bq/kg for each nuclide. Using this model, an annual effective dose of 1mSv per year corresponds to an activity index of 1.

This calculated index of 1 (~1mSv/year effective dose) presented by Markkanen (1995) is derived through a model, taking several factors into account, such as exposure time to habitant, radiation reaching the tissues and the organs in the body, attenuation coefficients of the material damping the radiation as well as thickness and density of materials. In light of the legislation of the European Commission council directive 89/106/EEC and EU´s Basic Safety Standards (2013) and the Construction Products Regulation (CPR) for EC-declaration of performance of construction materials, the assumption is that also Swedish industry is forced to accommodate to the criterion set out by the European Commission (EC).

In this article, the I-index will be addressed with a special focus on concrete. Using a field gamma ray spectrometer (portable hand-held spectrometer), RS 230 manufactured by Radiation Solutions Inc., an approach is made to correctly evaluate the I-index as suggested by the EC (1999). Comparisons are made with two laboratory procedures in order to evaluate the possible use of a hand-held spectrometer for correct evaluation of the I-index. Part of the study has also been to address I-indices that could be expected in concrete, by use of different crushed aggregates from the Swedish bedrock.

The study included crushed rock aggregates, which entail almost the full range of variations in natural radiation from gneisses and granitoids known within Sweden. The crushed aggregates were chosen from regions and suburbs encompassing Stockholm and Gothenburg, as well as the geographical region between these two major cities. A pre-study was done, making use of data from the Swedish geological survey, as to ensure that some materials would generate high natural radioactivity from the bedrock, as well as in some cases quite limited radiation. Ten different aggregates have been used from different quarries.

Earlier works within Sweden have focused on the evaluation of the effective dose received by habitants living in houses, detached houses and apartments (Almgren et al., 2008; Mjönes, 1986; Hultqvist, 1956). Almgren et al. (2008) used an approach, where habitants wore personal dosimeters for several weeks. Mjönes (1986) made thorough investigations of gamma radiation within Swedish dwellings and both Möre (1985) and Möre & Hagberg (1978) made dense analyses investigating both bedrocks and different concretes as construction materials. Some of their investigations could still be used to convert the observed levels of naturally occurring radiation in materials to I-index.

Assessment of natural ionizing radiation of building materials has also been presented by Rizzo et al. (2001) where different bedrocks and components used in building materials within Sicily (Italy) were assessed. Materials such as limestone, marble, cement, schists and other constituents that may be part of the building material were evaluated. Stoulos et al. (2003) made thorough analysis of some concrete mixes and granite materials used as tiles, as well as cements and different gravels aimed for building materials. The focus was on assessing materials used within Greece. In a recent publication, Stals et al. (2014) made several analyses with materials aimed for use in dwellings and suggest an additional approach to calculate the gamma dose rate, presented as I-index, by use of a portable equipment. The investigation, totaling hundred twenty six different analyses, where several samples were sent for external laboratory evaluation for comparative purposes, support their presented data of using a portable equipment. Eleven analyses were performed on concrete samples.

As a construction material, concrete is a mix of several components: aggregates, cement, water, filler (often limestone), additives and fly ash (optional). Aggregates as a constituent of concrete make up roughly 75% (weight) of the total mass (Neville & Brooks, 2010). Throughout the last two decades there has, within Sweden, been an increased use of crushed bedrock/rock, in order to preserve natural gravel/fluvial sediments reservoirs (Göransson, 2011).

2. METHODS

2.1 Concrete mixes

Ten concrete slabs were cast in the Swedish Cement and Concrete Research Institute´s laboratory. Each concrete slab is 150 mm in thickness and the width and length are 1.5 m. The aggregates used for the concrete mixes are in agreement with the suggested sieve curve for concrete repairs (EN 1766). All but one aggregate fulfilled the requirements. The anomaly is due to one single fraction, 0/18, which has been crushed coarsely by the entrepreneur sending the aggregates to CBI. No further crushing has been performed by CBI. All other investigated aggregates had been more thoroughly crushed by the entrepreneurs before sending the aggregates to CBI. For most aggregates, two or three sortings (0/4, 4/8 and 8/16) have been used and blended in accordance with the suggested sieve curve (EN 1766).

The water/cement ratio for all ten concrete slabs was set to 0.45. A CEM II/A-LL 42.5 R (Portland clinker cement with a portion of 6-20 % limestone and rapid hardening properties) from the Skövde cement factory was used for every concrete slab in order to limit the effect of other constituents than aggregates contributing to the natural ionizing radiation. The crushed aggregates contain two metasedimentary gneisses, one gneiss, two foliated grantioids (granitic gneisses), four granitoids with a more equigranular character and one gabbro/diabase.

2.2 Measurement procedure

The concept and basic method for evaluation of the total gamma rate, as well as the natural radioactivity of ^{40}K, ^{226}Ra and ^{232}Th relies on measurements attained using a field spectrometer (portable hand-held spectrometer), supplied by Radiations Solutions Inc. Measurements by the spectrometer on each concrete slab was executed approximately one month after casting. The configuration for measurements is called “2π configuration”, meaning that measurements are made on a single plane, with no other influencing factors from the side and top except cosmic and terrestrial background radiation.

Measurements have been registered from the centre, twice by rotating the spectrometer perpendicularly to the first reading, and 200 and 400 mm out from the centre of the cast concrete in four different directions, thus yielding a total of ten separate measurements on each concrete slab. Every measurement lasted four minutes (see Figure 1a,b).

The resolution, full width half maximum (FWHM) was predominantly between 4 and 5 % measuring the peak area of ^{232}Th. An approximate relative humidity (RH) on cut concrete slabs was executed shortly (5-6 weeks) after measurements ended on each concrete slab. The measurements imply an interval of 90-95 % in RH for each slab during sampling.

As aggregates and other components may not be perfectly homogeneously distributed in the concrete, evaluation of the measurements have been based on mean values of dose rate (μSv/h) and specific activities (Bq/kg), rather than results from individual measurements. The mean values are presented in Table 2.

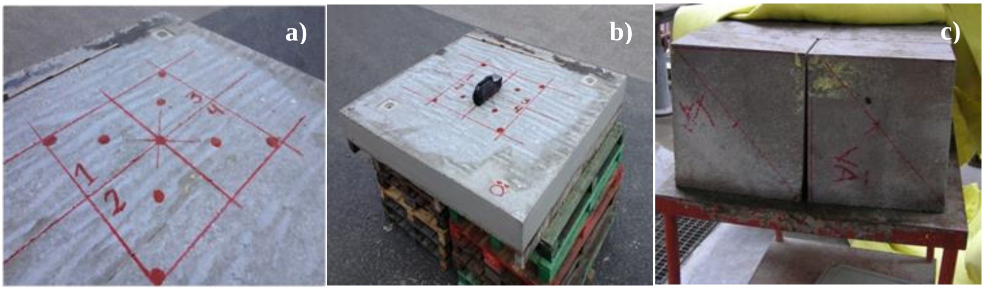


Figure 1. a) Square grids have been drawn (in red) on the concrete in order to simplify measurements and positioning of the instrument, 200 and 400 mm away from the centre in four directions. Red dots mark approximately the measurement points. **b)** Measurements on concrete slab using RS 230. The cast concrete slab is 0.15 × 1.5 ×1.5 m in thickness, length and width, respectively. The RS 230 measuring on the center point of the cast concrete. The concrete cast is supported by wooden pallets. **c)** The sawn/cut samples (0.3 × 0.3 m in length and width) represented in Figure 1a (grid squares) on a supporting rig. The samples are used for RH and comparative analyses.

2.3 Calibration and recorded quantities by the RS 230

The RS 230 was calibrated in a horizontal position by use of the calibration pads maintained by the Swedish geological survey. The pads are situated in Borlänge, Sweden and are used for calibration of air borne radiation survey equipment as well as portable equipment. The pads are approximately 11 m × 11 m × 0.5 m in size.

The RS 230 records the radioelement concentrations of potassium (K) in %. For radium (Ra) as well as thorium (Th) the concentrations are reported in equivalent ppm (parts per million). RS-230 measures gamma radiation energies from the decays of ^{40}K (1461 keV), ^{214}Bi (1760 keV) and ^{208}Tl (2615 keV), in three different energy windows, 1370-1570 keV, 1660-1860 keV and 2410-2810 keV respectively. The conversions from the measured radionuclides to total concentrations of potassium, uranium and thorium are based on natural occurrences of the isotopes (e.g. ^{40}K constitutes 0.0117 % of natural potassium). Moreover, in order to calculate concentrations of ^{226}Ra, ^{238}U and ^{232}Th from the daughter isotopes ^{214}Bi and ^{208}Tl, radioactive equilibrium in the decay chains is assumed (i.e. no element has been added or removed). In case of radioactive equilibrium, the activity concentration of all members of the decay chain is equal.

Hence, ^{214}Bi can be taken as a proxy for both ^{238}U and ^{226}Ra and ^{208}Tl could be taken as a proxy for ^{232}Th.

When assessing the data, the conversion of elemental concentrations to specific activity (Bq/kg) for each element follows the recommendations of IAEA (1989). The conversion factors used for transformation of elemental concentration to Bq/kg follows the conversion factors presented in Table 1.

Table 1. Conversion of radioelement concentration to specific activity (IAEA, 1989).

1 % Potassium (K)	=	313 Bq/kg	^{40}K
1 ppm Uranium (U, Ra)	=	12.35 Bq/kg	^{238}U, or ^{226}Ra
1 ppm Thorium (Th)	=	4.06 Bq/kg	^{232}Th

The RS 230 presents options to show dose rates in its display window. Usually, dose rate describes absorbed dose rate in air, which is presented in the unit nGy/h. However, within this article the dose rates measured by the RS 230 are presented as effective dose rate (µSv/h).

The instrument uses a 103 cm^3 Bismuth Germanium Oxide (BGO) crystal detector, with better efficiency than a similar sized NaI-crystal. The instrument set up is a 1024 channel spectra and an energy range of 30-3000 keV. The standard deviation is < 7 % (Löfberg, 2013).

2.4 Uncertainty of measurements on the concrete slabs

A minor study was conducted by Svensson (2014) in order to calculate the reproducibility of the measurements at one point, investigating the influence of distance from the centre of the concrete slab, as well as the potential influence of variation in results, by shifting directions of the instrument on the concrete slab. The influence or uncertainty of each nuclide (^{40}K, ^{226}Ra and ^{232}Th) were also investigated.

In short, an uncertainty of 10 % is validated (with a 95 % confidence interval). The influence of measuring with increasing distance from the centre of the slab, could be calculated. Considering a 95 % confidence interval, the uncertainty or possible loss of energy 400 mm away from the centre of the cast slab could be restricted to less than 5 %. No significant influence could be detected by rotating the instrument in different directions.

All in all, a maximum uncertainty (with a 95 % confidence interval), including a minor influence of shifting positions with the instrument across the slab are in the range of 10 %. Only marginal differences in uncertainty could be established between the three nuclides (^{40}K, ^{226}Ra, ^{232}Th) investigated.

2.5 Natural variations and background influence

The background variation including both cosmic and terrestrial background was checked each time prior to commencement of measurements on the concrete casts. The instrument was placed on supporting wooden pallets approximately 1 m above ground. The background influence was in the range of 0.065-0.071 µSv/h.

Part of the study was to investigate if the terrestrial radiation (radiation from ground level), would contribute to the total energy measured, due to the limited thickness of the concrete slabs. The background influence (terrestrial) was checked by use of sample 3 (concrete slab), which yielded an I-index > 1. Comparison was made by measuring on the cast concrete, with a steel container, approximately 1.5 × 1.5 × 1.2 m in height and filled with 1 m of water, placed underneath the concrete slab. Thereafter the steel container was removed and measurements were made on the cast concrete resting on wooden pallets. Two measurements were made at the centre of the slab and a mean value was calculated for each scenario. The increase (having the steel container removed), in relation to the I-index, was 1.2 %.

2.6 Comparative analyses for control and validation of results presented by CBI

Correlation of the specific activity (Bq/kg) for ^{40}K, ^{226}Ra, and ^{232}Th between the different procedures (CBI measurements and laboratory methods) have been achieved by sawing each of the cast concrete slabs in four smaller concrete blocks (see Figure 1c), where each block represent at least three readings from the measurements on the full-scale cast slab. One of the four blocks from each cast slab was selected for laboratory analyses. The selected concrete block were sawn into even smaller blocks and finally crushed with a small laboratory jaw crusher (Morgårdshammar).

Approximately 30 kg were crushed from each cast. Secondary crushing was performed with a laboratory gyrocrusher, adjusted for a maximum crush size of 6 mm. Most crushed concrete materials were hereby < 6 mm in fraction before being sent for analysis. Thereafter, each sample was reduced in accordance with SS-EN 932-2 (1999). Finally, two subsamples, approximately 3-4 kg each was sent to STUK as well as to CRPG/CNRS. An assumption is made, by use of the different crushing stages and reduction of the larger sample into sub-samples, that the separate samples are more or less homogenous.

2.7 Laboratory measurements of the concrete samples, STUK and CRPG/CNRS

2.7.1 STUK

The Radiation and Nuclear Safety Authority of Finland (STUK) uses advanced gamma spectrometric analyses, which is an accredited testing method - gamma-spectrometry, in-house guide VALO 4.5 (Klemola et al., 2010). The gamma lines for assessing the three radioactive nuclides are for ^{40}K -1460.8 keV, ^{226}Ra - 186.2 keV (contribution of Uranium-235 subtracted) and for ^{232}Th, the decay of ^{228}Ac at 911.2 keV.

The activity concentration of gamma-emitting nuclides are measured with a low-background, high-resolution HPGe spectrometer. The detectors are placed in cylindrical background shields made of 120–140 mm thick lead and lined inside with cadmium and copper. The measured energy range is 30–2700 keV. The relative efficiency of the detectors varies from 37 to 90 % and energy resolution between 1.6 keV - 2.1 keV at 1.33 MeV. The samples are measured either in 35 ml or 105 ml cylindrical beakers, or in 0.5 litre standard Marinelli-beaker.

The correction for sample height and density, as well as the effect of true coincidence summing, is taken into account in the calculation of the results. The uncertainties include both statistical uncertainty and uncertainty due to the efficiency calibration.

The uncertainty of results (2σ) with a 95% confidence interval, is approximately 9-10 % determined from a mean value of all received calculations, irrespectively of which energy field being measured (^{40}K, ^{226}Ra or ^{232}Th).

2.7.2 CRPG/CNRS

Centre de Recherches Pétrographique et Géochimiques/ Le Centre National de la Recherche Scientifique (CRPG/CNRS) uses Inductively Coupled Plasma/Optical Emission Spectrometry (ICP-OES) to measure major and Rare Earth Elements elements. The technique relies on that excited atoms and ions in a discharge plasma create a unique emission spectrum specific to each element.

The results for potassium (K) have been reported as potassium oxide (K_2O). A correction for the potassium (K), making up 83 % of the total weight of the K2O molecule has been applied in order to calculate the mass activity in Bq/kg of 40K (see Table 2). CRPG/CNRS make us of an ICP-OES called Thermo Icap 6500 with a radial torch.

For determination of the Rare Earth Elements, such as thorium and uranium, CRPG/CNRS uses Flow Injection and Low Pressure On-Line Liquid

Chromatography Coupled to ICP-MS. The instrument in use at CRPG/CNRS is a Thermo X7.

The sample is divided several times in order to have a suitable sample size. At the end, approximately 300 mg are used for further processing. The methodology procedure is explained in detail by Carignan et al., 2001.

3. RESULTS

The range of values (Table 2) for the effective dose rate measured on the concrete slabs varies from a maximum of 0.25µSv/h to 0.08 µSv/h with an average of 0.14µSv/h. The average of 0.14 µSv/h corresponds to ~0.9 in I-index for CBI measurements (see Table 2, concrete sample 4) and approximately 1 in I-index, compared with the other analytical methods.

Table 2. The mean specific activity (Bq/kg) of ^{40}K, ^{226}Ra and ^{232}Th of ten separate measurements on each cast concrete slab measured by CBI. The table also shows results from STUK and CRPG/CRSN and their related I-indices. The effective dose rate (µSv/h) is presented for measurements performed by CBI.

Concrete samples	Bedrock	Specific activity (Bq/kg) from results obtained by CBI			Activity-value, I (CBI)	Effective dose rate (µSv/h)	Specific activity (Bq/kg) from results obtained by STUK			Activity-value I - (STUK)	Specific activity (Bq/kg) from results obtained by CRPG/CNRS			Activity-value, I (CRPG/CNRS)
		^{40}K	^{226}Ra	^{232}Th			^{40}K	^{226}Ra	^{232}Th		^{40}K	^{226}Ra	^{232}Th	
1	Granitoid	1134.6	157.8	134.9	**1.58**	0.25	1220.0	250.0	160.0	**2.04**	1176.1	233.7	146.8	**1.9**
2	Granitoid	1216.2	92.3	168.5	**1.56**	0.25	1230.0	100.0	210.0	**1.79**	1294.5	124.9	222.1	**1.96**
3	Granitoid	1283.3	118.7	87.7	**1.26**	0.2	1460.0	150.0	100.0	**1.49**	1400.8	113.1	87.9	**1.28**
4	Granitoid/Gneiss	748.1	53.7	92.6	0.89	0.14	790.0	62.0	110.0	**1.02**	700.4	46.5	119.0	0.98
5	Gneiss (metasedimentary)	983.3	26.1	47.2	0.65	0.1	920.0	41.0	42.0	0.65	932.9	35.1	39.9	0.63
6	Gneiss (metasedimentary)	871.2	42.7	42.5	0.65	0.1	823.0	36.0	41.0	0.6	895.0	26.9	31.4	0.54
7	Gneiss	852.3	45.3	39.2	0.63	0.1	1000.0	15.0	41.0	0.59	1030.1	15.7	39.3	0.59
8	Granitoid/Gneiss	704.3	42.5	31.6	0.53	0.08	690.0	38.0	29.0	0.5	632.3	27.3	28.9	0.45
9	Granitoid	606.1	39.6	31.1	0.49	0.08	557.0	36.0	27.0	0.44	571.3	28.3	18.0	0.37
10	Gabbro	381.9	22,00	15.3	0.28	0.04	261.0	8.2	7.3	0.15	282.4	7.4	7.1	0.15

For the I-index, the maximum value is 1.6 (2.04)[1], (1.90)[2] with a minimum of 0.5 (0.15)[1], (0.15)[2] and an average value of 0.9. The three samples yielding the highest values show results in the interval I = 1.3-1.6 (1.5-2.0)[1], (1.3-1.9)[2] . Comparing CBI results with results from STUK, an additional sample reaches the acceptance

[1] I-index value calculated from the specific activity of ^{40}K, ^{226}Ra and ^{232}Th results received from STUK, calculated from part of the same cast concrete material in a crushed and dried state.

[2] I-index values obtained from calculation of the specific activity for ^{40}K, ^{226}Ra and ^{232}Th for results received in elemental concentrations of K, U and Th from part of the same concrete material as measured by CBI.

criterion (>1), albeit a border liner. Generated results from STUK as well as calculated specific activity values (Bq/kg) from CRPG/CNRS show a fairly good correlation in the medium to lower intervals (I-index 0,5-1). At a level of ~1 in I-index for STUK and CRPG, the CBI measurement (sample 4) apparently underestimates the I-index by approximately 10 %. A comparison of the calculated I-indices from the ten concrete samples, with its corresponding I-values from CBI, STUK and CRPG/CNRS, is presented in Figure 2.

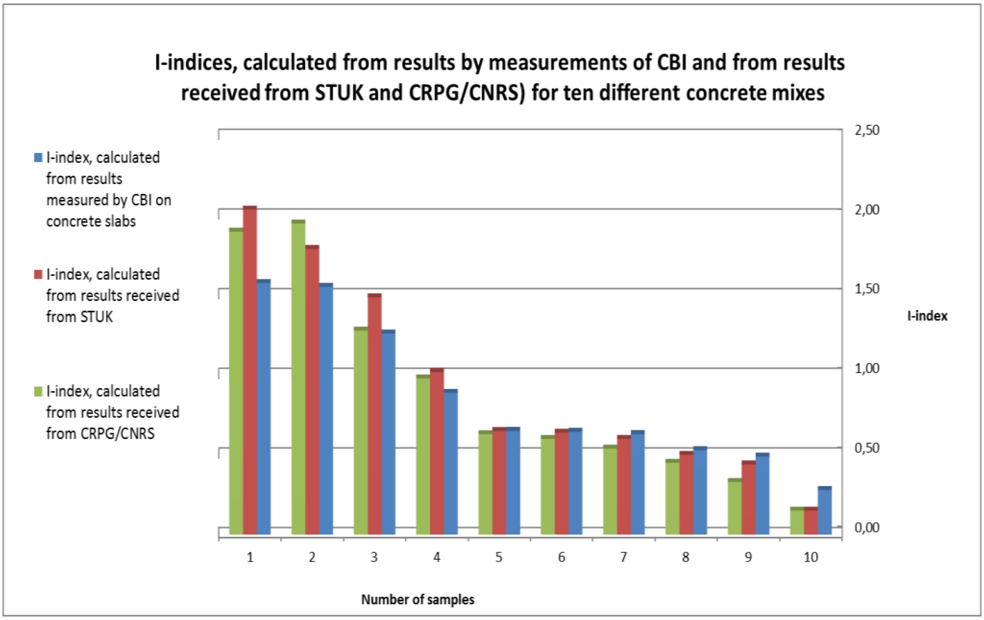


Figure 2. Calculated I-indices from results obtained from CBI (portable gamma ray spectrometer), STUK (laboratory gamma ray spectrometry) and geochemical analysis (CRPG/CNRS) of the ten concrete mixes.

The results in specific activity (Bq/kg) of the samples vary considerably between the laboratories, when an I-index > 1 is calculated (sample, 1, 2, 3, Table 2, figure 2). There are obvious differences in measured specific activity (Bq/kg) depending on procedure used. This is also noticeable for sample 4 (Figure 2). where the measurements of CBI are slightly lower than results obtained from STUK and CRPG/CNRS. In order to evaluate this discrepancy, each specific nuclide (^{40}K, ^{226}Ra and ^{232}Th) were plotted separately to compare the results between CBI and the laboratories (Figure 3, 4, 5).

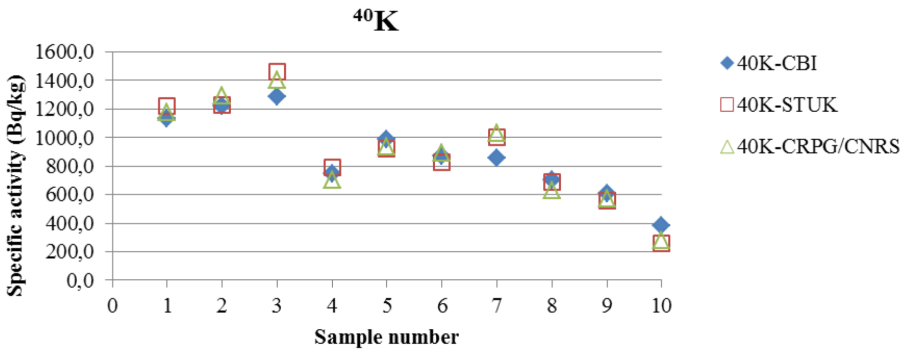


Figure 3. Comparative calculations of measurements for ^{40}K.

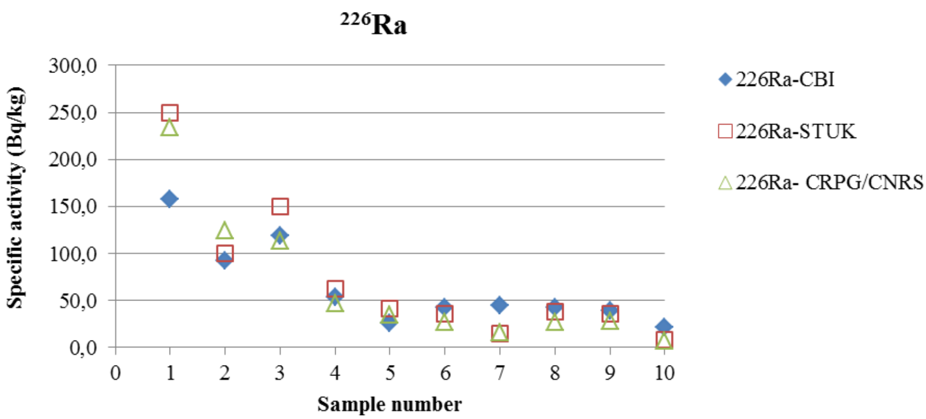


Figure 4. Comparative calculations of measurements for ^{226}Ra.

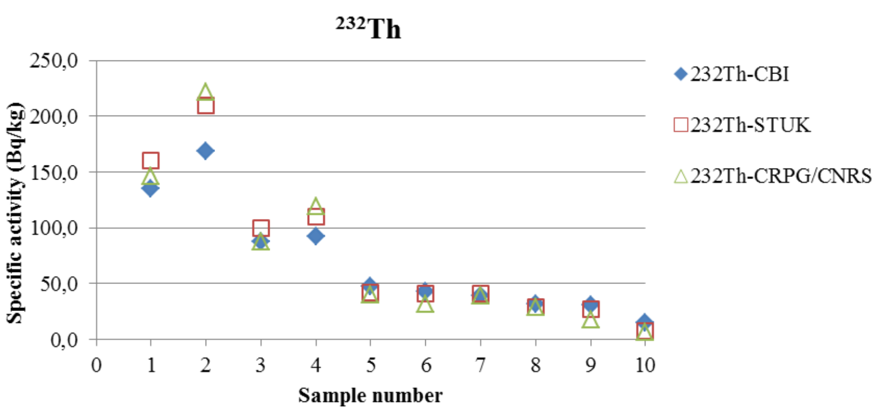


Figure 5. Comparative calculations of measurements for ^{232}Th.

The results reveal that for ^{40}K (Figure 3), there is a slight discrepancy for samples 3, 7 and 10, but on the whole there is a good agreement between the different procedures.

However, for ^{226}Ra, there is a large discrepancy observed for in particular sample 1 between CBI- measurements and the two laboratory methods. A similar scenario could be seen for ^{232}Th for sample two, which is strongly underestimated in the CBI measurement compared to the laboratory methods, which in general show a better consensus. The largest deviation occurs, when the highest level of ^{226}Ra and ^{232}Th is recorded by the laboratory methods, which is also true for ^{40}K (sample 3).

Concerning I-index > 1, there is seemingly an increased gap in calculated values between CBI values of I-index and the laboratory methods. For I > 1, the underestimation by CBI measurements is ~15-25 %.

On the other end, for concrete samples yielding values with an I-index < 0.5, the results in Figure 2 show that the CBI measurements slightly overestimate the I-index compared to laboratory procedures. The same trend is also seen in Figure 3, 4 and 5 (sample 8, 9 and 10) where each nuclide seemingly contributes with a slight excess of energy compared to laboratory methods, particularly in relation to results from CRPG/CNRS.

The contribution of each nuclides contribution to the I-index were further assessed. Specific interests are drawn to the samples with an I-index > 1 (sample 1, 2, 3 and 4). Table 3 summarizes the contribution of each nuclides fraction to the I-index.

Table 3. The table shows the contribution of each nuclide (^{40}K, ^{226}Ra, ^{232}Th) fraction to the I-index for the ten concrete samples investigated. Comparative data are presented for the three investigated methods/procedures.

Contribution to the I-index for each nuclide (^{40}K, ^{226}Ra and ^{232}Th)												
	Measurements by CBI				Measurements by STUK				Measurements by CRPG/CNRS		
Concrete samples	**^{40}K**	**^{226}Ra**	**^{232}Th**		**^{40}K**	**^{226}Ra**	**^{232}Th**		**^{40}K**	**^{226}Ra**	**^{232}Th**
1	0.38	0.53	**0.67**		0.41	**0.83**	**0,80**		0.39	**0.78**	0.73
2	0.41	0.31	**0.84**		0.41	0.33	**1.05**		0.43	0.42	**1.11**
3	0.43	0.40	**0.44**		0.49	**0.50**	**0.50**		**0.47**	0.38	0.44
4	0.25	0.18	**0.46**		0.26	0.21	**0.55**		0.23	0.16	**0.60**
5	0.33	0.09	0.24		0.31	0.14	0.21		0.31	0.12	0.20
6	0.29	0.14	0,21		0.27	0.12	0.21		0.30	0.09	0.16
7	0.28	0.15	0.20		0.33	0.05	0.21		0.34	0.05	0.20
8	0.23	0.14	0.16		0.23	0.13	0.15		0.21	0.09	0.14
9	0.20	0.13	0.16		0.19	0.12	0.14		0.19	0.09	0.09
10	0.13	0.07	0.08		0.09	0.03	0.04		0.09	0.02	0.04

According to the CBI.-results, ^{232}Th gives the largest contribution to the gamma-index in all four samples with activity index higher or close to 1, whereas in both laboratory methods, ^{226}Ra contributes the most in sample 1, and in the CPRG/CNRS results, ^{40}K makes the major contribution to the activity index in sample 3.The fraction which contributes mostly to the I-index is marked in bold letters. The fractions of each nuclide from CRPG/CNRS are similar to the values of STUK and supports that all nuclides contribute in part substantially even at higher I-indices.

Further analyses was also executed were regression analyses were used to evaluate the calculated I-indices of each method/procedure. Accordingly plots of the I-indices where made between results of STUK and CBI as well as for I-Indices between CRPG/CNRS and CBI. The results are presented in Figure 6.

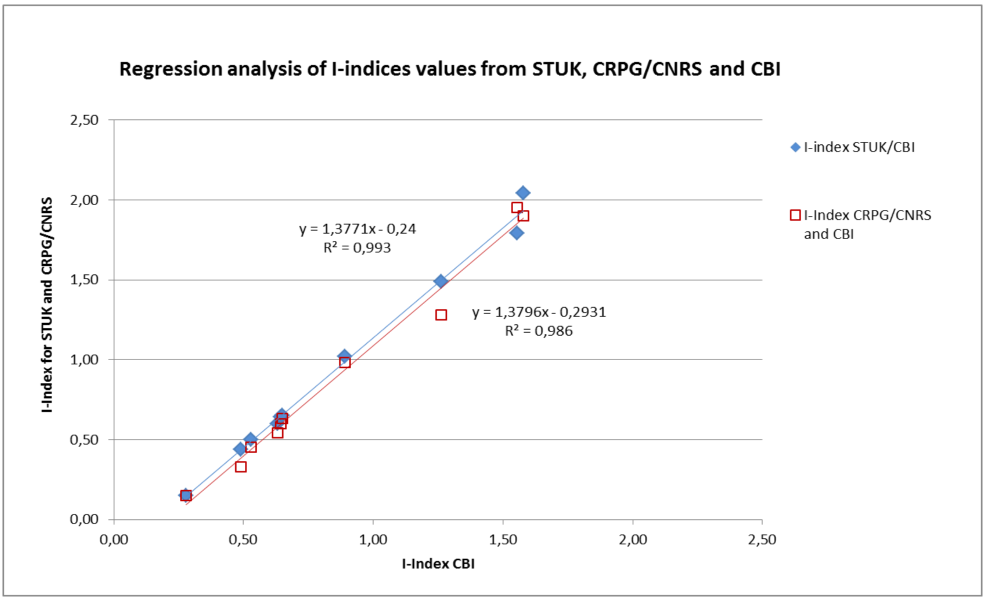


Figure 6. Correlation between the calculated I-indices by STUK and CBI as well as the calculated I-indices of CRPG/CNRS and CBI. A linear relationship could be suggested for both cases.

4. ANALYSIS

The regression analyses performed on the ten samples yielded two solid correlations with $R^2 \sim 0.99$ in each case, where a linear correlation coefficient could be established. This implies that a linear approach to correlate between the calculated I-indices is legitimate. The good agreement further suggests that knowing the conditions, the portable spectrometer can be used under the specified conditions and a correct estimate of the I-index can be achieved by use of the above correlation formulas.

However the differences in results between CBI and the analytical laboratories are of interest. The differences shown for sample 1, 2, 3 presented in Figure 4 and 5 (^{226}Ra and ^{232}Th) could in part be due to the fact that laboratory samples are dried and crushed, and hence the influence of humidity in the sample is lowered. The humidity (RH) was 90-95 %, when measurements were performed by CBI, which would most likely dampen the ionizing radiation released by the concrete slabs (IAEA, 2003). This may explain that the discrepancies between some samples are more pronounced at higher specific activities of the nuclides. Stals et. al (2014), who used an equipment, which could be used in an 2π and 4π configuration encountered a similar loss of energy for ^{226}Ra. The authors proposed that the loss could be due to partial loss of radon (when measuring in an open space) resulting in a lower measured radium progeny concentration as reported by Al-Jarallah et al., 2005. This will not likely occur in an analytical environment using sealed containers. This may also explain the large discrepancy in measurements for sample 1.

However, the loss of energy at higher energy peaks could also be due to the thickness of the concrete slabs. Considering sample thickness not all energy could be accounted for at a thickness of 150 mm (IAEA, 2003), which has been demonstrated by Grasty (1987). By use of a NaI-crystal approximately ~90 % of the energy of the nuclide ^{232}Th, could be accounted for having a thickness of 150 mm. The yield for ^{226}Ra and ^{40}K will most likely be higher, due to their lower energy gamma rays. This may in part explain why ^{232}Th is underestimated in the CBI-measurements at higher elemental concentrations (Figure 5).

Thirdly, there is most likely a minor loss of energy (radiation), due to the concrete sample size, since these are not infinite in size and the results presented are a mean value from every measurement on the cast slab. This is also in line, with calculations by Svensson (2014) demonstrating a minor loss of energy with increasing distance from the centre of the slab.

Accounting for the discrepancies at the lower values of the I-indices, see Figure 2 (sample, 6-10) CBI measures higher values than the calculated I-indices of STUK and CRPG/CNRS. The difference is likely due to background influence: The background influence is normally subtracted, when analyzed at a laboratory (STUK). The background influence, should however be small for I-values > 1 due to the tests performed by CBI, but when the mass activities of the three nuclides are fairly low (I-index <0.5), it seemingly contributes slightly to the field measurements.

The analytical laboratory procedure of calculating the gamma-emitting nuclides is to be viewed as the primary methodology to evaluate the ionizing radiation from ^{40}K, ^{226}U, ^{232}Th, due to the controlled conditions, where the influence of background radiation is reduced and humidity and sample size are according to a standardized procedure. Also, the buildup process of ^{222}Rn as shown by Mauring and Gäfvert (2013), using sealed containers seemingly obtain reliable results. The variability in humidity, cosmic radiation and terrestrial background varies for measurements in the field and could all affect the results when using a portable hand-held spectrometer (IAEA, 2003).

Furthermore, the analytical laboratory methodology used by STUK (VALO 4.5) is in line with the harmonized methodology developed by the European Union in 2013, as how to measure gamma radiation from construction materials. The geochemical analytical procedure using ICP OES/MS is sensitive in its character, since only a very limited amount is evaluated from a large bulk sample, meaning that the samples need to be very homogenous to avoid anomalies.

Investigations made have shown that an empirical approach by use of a hand-held spectrometer, measuring on cast concrete, with fixed size and thickness, could be performed with accuracy at intermediate I-values, ranging from 0.5-1. Some areas where this could be applied would be within the precast concrete industries, where similar measurements could be obtained, with known concrete thicknesses.

For materials exceeding the stipulated levels of 1 mSv/year, an approach of a separate dose assessment is recommended by the EU-commission Construction Product Regulation (CPR) and its related Basic Safety Standards (BSS), taking into consideration the full extent of its use within the dwelling (EC, 2013). A separate dose assessment approach is under review by the EC technical committee 351/WG 3/TG 32 and a suggestion will most likely be presented during 2014-2015.

5. CONCLUSIONS

The results indicate that at least six samples of the investigated ten concrete samples are in agreement with the stipulated levels set out by EC (1999, 2013). The performed measurements on the concrete slabs indicate that some concrete mixes could have an I-index ~2, which is twice the recommended level.

It is often the granitoids which display high natural radiation. The cause for the elevated levels of ionizing radiation is mainly due to moderate to high levels of ^{232}Th in the concrete mixes containing granitic material, even though ^{226}Ra and ^{40}K in some cases contribute substantially to the I-index.

The investigations have further shown, that there are some limitations to correctly evaluate the total contribution of ionizing radiation of ^{40}K, ^{226}Ra and ^{232}Th from a portable hand-held spectrometer, due to practicalities of sample size, thickness and external factors, such as humidity and background variations. However, the regression analysis presented within this paper illustrates that there are good linear correlations with the comparative laboratory methods. As such, the methodology proposed by CBI could be suggested, to roughly estimate the I-index, knowing the thickness and recipe of the concrete.

The use of the portable hand held spectrometer further revealed that a total effective dose rate of ~0,14 µSv/h could be estimated as being equal to 1 in I-index.

The calculations as presented within this paper have to be regarded as estimates, representing 150 mm thick concrete. An increase in ionizing radiation from concrete with increased thickness is to be expected.

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