Ionizing Radiation in Concrete and Concrete Buildings – Empirical Assessment

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Preface

This licentiate work was carried out at the Swedish Cement and Concrete Research Institute (CBI) in Borås and at Royal Institute of Technology (KTH) in Stockholm. The licentiate work was initiated in February 2012 and was finished in August 2016.

The project was financed by the Swedish Consortium on Financing Basic Research in the Concrete Field.

The project had a reference group with people from the different members of the Swedish Consortium as well as tutors representing the Royal Institute of Stockholm (KTH), department of Civil and Architectural Engineering, division of Concrete Structures (Johan Silfwerbrand), the Swedish Geological Survey (Cecilia Jelinek), the Swedish Cement and Concrete Research Institute (Jan Trägårdh) and department of Radiation Physics, Sahlgrenska Academy, University of Gothenburg (Mats Isaksson). Among the Swedish Consortium members in the reference group participated Kent Slade (Strängbetong), Monica Soldinger Almefelt (Peab/Swerock AB) and in part Mats Karlsson and associates (Färdig Betong AB).

The author is very grateful to my supervisors and especially Professor Johan Silfwerbrand, who always tried to make time for discussion, evaluation and fruitful guiding in the process to finalize the thesis. Many thanks to my supervisor Cecilia Jelinek, Ph. D., at the Swedish Geological Survey, who contributed with great quality in the discussions in the reference group and by improving the Papers produced. Many thanks as well to my supervisor Professor Mats Isaksson, who gave me the necessary means to yet further understand the issues of radiation. Finally, special thanks to Jan Trägårdh, Ph. Lic., who contributed with valuable discussions and support when needed.

A passionate thought is also addressed to my colleagues at Borås, who helped me to produce all the concrete specimens necessary during the project.

Also, I address sincere thanks to Eva Lundgren at CBI, Stockholm, who came to my rescue, when literature references seemed impossible to get a hand on.

Göteborg. August 2016

Magnus Döse
Summary

One of the major issues with radiation from the natural isotopes $^{40}$K, $^{226}$Ra ($^{238}$U) and $^{232}$Th and their decay products is the forthcoming legislation from the European Commission in relation to its Basic Safety Directive (2014). The European legislation is mandatory and could not be overthrown by national legislation. Hence, even though the BSS is still a directive it is foreseen as becoming a regulation in due time.

The reference value of the natural isotopes, from a radiation point of view, set for building materials is 1 mSv per year (EC, 2014). Earlier recommendations (The Radiation Protection Authorities in Denmark, Finland, Iceland, Norway and Sweden, 2000) within the Nordic countries set an upper limit at 2 mSv per year of radiation from building materials.

The main objective within the frame of the thesis was to investigate gamma radiation in relation to Swedish aggregates and their use as final construction products and the applicability and use of a model (EC, 1999) for building materials to calculate the effective dose within a pre-defined room. Part of the thesis also investigates different methodologies that can be used to assess the radiation in a construction material made up of several constituents (building materials) and aims to show that for some purposes as for the construction industries (precast concrete), that a hand-held spectrometer can be used with good accuracy, even though the object is limited in thickness and size. Secondly, the author proposes a simplified way of assessing the radiation in a construction material by use of correlation coefficient of a specified recipe by use of a hand-held spectrometer. Moreover, an understanding of the different building materials’ contribution to the finalized construction product, e.g. concrete is demonstrated, and how to achieve a good control of the radiation levels in the concrete building.

A second part of the thesis comprised the correlation of the decay chain of uranium ($^{226}$Ra) and the contribution of radon gas ($^{222}$Rn) from the finalized construction product, concrete. The thesis has focused on the release of $^{222}$Rn in relation to the change in relative humidity (RH) during hydration. This process has been monitored and two different set ups were deployed. Water cement-ratios of 0.45 and 0.65 were used for thirteen different samples measuring 300 × 300 × 150 mm or 300 × 300 × 200 mm.

The methodologies to measure the specific activity (Bq/kg) of the natural radionuclides ($^{40}$K, $^{226}$Ra $^{232}$Th) are primarily assessed in two different ways. Firstly, by using a hand-held spectrometer, where the readings are given in elemental concentration (weight %) of the natural radionuclides and converted to specific activity (Bq/kg). These analyses are complemented by gamma spectrometric measurements using semiconductors with High Purity Germanium Detectors (HPGE) given the reading as specific activity (Bq/kg). The use of a hand-held spectrometer also gives readings of the dose rate as ambient dose rate, $H_{a}(10)$, as a comparative tool to the elemental concentrations of each radionuclide. Thirdly, a comparative analysis (Paper I) has also employed geochemical analysis of the natural radionuclides using ICPMS/Eos to assess the elemental concentrations (%) of the radionuclides in the different concrete samples.

For radon assessments a methodology (ionizing pulsation chamber) using the decay rate of alpha energies from $^{222}$Rn and $^{218}$Po is employed. The amount of decay per unit time is calibrated in relation to a well-defined radon gas level. The readings or the output from the radon gas monitor are then displayed as $^{222}$Rn content in air in the unit Bq/m$^3$. 
The results within the scope of the project are multifold, but could be summarized as:

1. A hand-held spectrometer has limitations due to its calibration but may well be used for radiation measurements on concrete products. However, a size of at least 1500 × 1500 mm is recommended. A thickness of at least 150 mm would give a result, which is fairly trustworthy for an I-index of 1 or below.

2. Aggregates as part of the concrete constitute the major building constituent (~70-80 wt. %) and as such, their content of radiation will be the dominating influence of the concrete’s final activity concentration (Bq/kg).

3. Using a recipe of 350 kg/m³ cement and a water-cement-ratio of 0.45 for an aggregate with an I-index of 1 yield an approximate corresponding effective dose rate of 0.65 for a room with dimensions 3×4×2.5 m and with 200 m thick concrete walls.

4. An aggregate with an I-index of 1.3-1.4 may well be used and still have an effective dose < 1 mSv per year if correct blending of aggregates are performed.

5. The calculated value of the final contribution of natural radioactivity within a concrete, using measured specific activities (Bq/kg) of each constituent as part of the concrete corresponds well with the measured specific activity of the final concrete.

6. Radon gas in relation to relative humidity (RH) seemingly has a low variation for a concrete with a w/c ratio of 0.45. For w/c ratios of 0.65 a clear trend of lower radon gas exhalation rates as RH decreases has been established. At RH levels below 65-70 % the radon gas levels seem to have a more stabilized progress, meaning no difference in radon gas exhalation could be established at RH levels below 65-70 % from the surface of the concretes.

Further research is recommended between the links of a product which is validated for Declaration of Performance (DoP) and its impact to the total effective dose within a finalized dwelling. Depending on layers used within a building no clear relation exists today and validations are necessary to check if the measured/calculated values of a construction product will retrieve the corresponding effective doses as foreseen by theoretical models. Secondly, the contribution of the radon gas to the air in relation to use of different alternative binders is recommended. A decrease of the radon gas from the concretes using a combination of binders or additives may give the concrete producers very good prerequisites to handle demands in public procurements where low levels of radon gas as part of environmental measures for indoors use are regulated.
**Sammanfattning**

En av de huvudsakliga frågeställningarna med strålning från de naturliga isotopeerna $^{40}$K, $^{226}$Ra ($^{238}$U) and $^{232}$Th och deras sönderfallsprodukter är den kommande lagstiftningen från Europeiska kommissionen i relation till EU:s direktiv för säkerhetsföreskrifter för miljö och hälsa (EC, 2014). Den europeiska lagstiftningen är prejudicerande och kan inte omkullkastas av nationell lagstiftning. Härav, även om Säkerhetsföreskrifter för miljö och hälsa är rekommendationer kommer det sannolikt så småningom att bli en förordning.

En del av licentiatuppsatsen har också undersökt olika metoder som kan användas för att mäta strålning i en färdig produkt, som består av många olika byggmaterial. För det första för att visa att under vissa förutsättningar, såsom för prefabricerade betongelement, kan en handburen spektrometer användas med god tillförlitlighet även om objektet är relativt litet. För det andra är föreslaget ett enkelt koncept för att utvärdera strålningshalten i betong med hjälp av konversionskoefficienter för ett recept där handburen spektrometer används. Tillsika visas hur man genom god förståelse av strålningshalten i olika byggmaterial kan ha god kontroll på strålningshalten i betongmaterialet.

**Metoderna som använts för att undersöka den naturliga strålningen från radionuklidera ($^{40}$K, $^{226}$Ra, $^{232}$Th) är i huvudsak två. Den första metoden är att använda en bärbar spektrometer, där mätresultaten presenteras som koncentrationer av radionuklida i vikt %, vilket sedan kan konverteras till specifik aktivitet (Bq/kg). Dessa analyser kompletteras med den andra metoden, gamma-pekemtrometri, där analyser är utförda i laboratorimiljö med hjälp av semikonduktorer, som använder High Purity Germanium Detektorer (HPGE), vilka ger mätvärden som specifik aktivitet (Bq/kg). Den bärbara spektrometern ger också miljöekvivalentdosraten, $H_*(10)$, vilket kan jämföras mot den uppmätta grundämneshalten av respektive radionuklid (vikt %). Dessutom har ytterligare analyser utförts med hjälp av geokemi och ICP MS/Eos, där innehållet av respektive radionuklids grundämneshalt har utvärderats i de olika betongproverna.

För utvärdering av radonhalten ($^{222}$Rn) har en metod använts (joniserande pulsationskammare), som utnyttjar alfasönderfallet från $^{222}$Rn och $^{218}$Po. Mängden sönderfall per tidsenhet är i proportion till en kalibrerad känd radongashalt. Information om radongashalten i luft kan utläsas i en display på instrumentet, där halten är angiven såsom $^{222}$Rn i luft i enheten Bq/m$^3$. 

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Resultaten inom projektet är relativt omfattande, men kan summeras såsom;

1. En handburen gammaspektrometer har begränsningar beroende av dess kalibrering och mot vilket underlag den är kalibrerad, men kan mycket väl användas för gammastrålningsmätning på betongprodukter. Men, en tillräcklig storlek är nödvändig för att erhålla pålitliga resultat. En dimension av minst $1500 \times 1500 \times 150$ mm i tjocklek torde ge resultat, vilket är tillräckligt pålitligt för korrekt utvärdering av Index 1 eller lägre.

2. Krossmaterial utgör huvuddelen av betong och således är dess gammastrålningssegenskaper mycket avgörande för den slutliga betongprodukterns strålninghalt.

3. Om man använder ett recept med $350$ kg/m$^3$ cement och ett vct av $0,45$ för ett krossmaterial med ett I-index av 1 ger ett motsvarande värde i effektiv dos $\sim 0.65$ mSv/år i en rumsvolym motsvarande $3\times4\times2,5$ m med en betontjocklek av $200$ mm utifrån utvärderad regressionsanaly.

4. Ett krossmaterial med ett I-index om $\sim1.3-1.4$ kan användas i en betongkonstruktion, där slutlig effektiv dos understiger $1$ mSv per år, förutsatt att korrekt proportionering av krossmaterial som används utförs.

5. De kalkylerade/ beräknade värdet av det slutliga bidraget av naturlig strålning från en betong, där respektive byggsammanlag, som utgjorde del i betongen, hade analyserats med erhållna värden av specifik aktivitet (Bq/kg) stämde väl mot den slutligt analyserade halten av specifik aktivitet i den färdiga betongen.

6. Radongas i relation till relativ fuktighet verkar ha en mindre betydande influens när betongens vct är $0,45$. För betong med ett vct av $0,65$ har dokumenterats en tydlig trend med sjunkande radongashalter med sjunkande relativ fuktighet. Vid RF-halter om $65-70$ % förefaller radongasexhalationen stabiliseras och ingen ytterligare förändring kan noteras vid lägre halter av relativ fuktighet.

Vidare rekommenderas avseende sambandet mellan mätning av byggprodukter avsedda för produkt-deklarering (DoP) och deras påverkan på den slutliga effektiva dosen i en färdig byggnad. Beroende av olika skikt inom en byggnad finns idag ingen tydlig korrelation och validering är nödvändig för att se om uppmätta värden av en byggprodukt kommer att erhålla den teoretiskt beräknade effektiva dosen enligt tillgängliga modeller. Även radongasen bidrag rekommenderas att undersökas i relation till alternativa bindemedel och tillsatsmaterial. En minskad avgång av radon med rätt kombination av olika bindemedel och tillsatsmaterial kan ge betongproducerer mycket goda förutsättningar att klara lågt ställda krav på radongashalter i betongprodukter och samtidigt uppfylla lågt ställda miljökrav för inomhusmiljön.
List of publications

The following papers are included in the Thesis


Authors: M. Döse, J. Silfwerbrand, C. Jelinek and J. Trägårdh


Author: M Döse


Authors: M. Döse, J. Silfwerbrand, C. Jelinek, J. Trägårdh and M. Isaksson

The planning, analyzing and writing were primarily performed by the main author. The co-authors have guided the work and contributed to papers I and III with comments, suggestions and amendments. Paper II was produced by the author alone and reviewed by external expertise as a contribution to an IAEA-congress held in Turin, 2014. Döse has performed most of the laboratory work, except the initial measurements of radon gas, and is responsible for the evaluation of all experimental data as well as calculations thereof.

Other publications by the author on the same topic


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1 Introduction

1.1 The issue with radiation

Natural radioactive decay of elements is a process, which is steadily ongoing. The natural decay of some elements is due to their unstable nature of the nucleus. A large excess of neutrons in relation to protons in order to keep the nucleus stable gives rise to a final collapse (Isaksson, 2011a), which initiates a break-down chain, which is not recognized by the normal eye.

The radioactive elements, often as part of different minerals, are in various degrees part of all sediments, volcanic and magmatic rocks on earth. Thus, glaciofluvial sand or gravel as a product of physical and chemical weathering as well as magmatic bedrock all contain the natural radioisotopes (40K, 238U and 232Th) in various degrees. They are a necessity as to ionize materia, so-called natural radiation.

The mere length of the break-down processes poses a dilemma. The uranium chain has a half-life of 4.5 million years (Fowler, 1996). A half-life is the time for the element to lose half of its radioactive content. Hence, it could be understood, that for those materials, which are used today, there will be no significant change in the activity of natural decay during our life-time or several life-times due to the very long time for natural decay of our elements to end. However, the forces of nature do inflict on the content of natural isotopes within materials, such as aggregates. Water, the most valuable source on earth, contributes in part to leach aggregates, by slow diffusion, through percolation in glaciofluvial deposits. This is not always considered, when using crushed rocks, where the percolation of water or hydraulic conductivity is most often very small in comparison to glaciofluvial deposits.

The naturally occurring radionuclides of interest are 40K, 226Ra (238U) and 232-Th, and their daughter isotopes. 40K constitutes 0.012 % of all natural potassium within the bedrock, and occurs in rock-forming minerals such as potassium feldspar (10-14 % K) and micas, 9-10 % K (Jelinek & Eliasson, 2015). Uranium and thorium, on the other hand, occur only as trace elements in rocks, mostly in accessory minerals, e.g. uraninite, allanite, apatite, titanite, xenotime (Jelinek & Eliasson, 2015).

In parts of Sweden, sediments such as black shales (alunskiffer) are enriched in uranium. It occurs in different parts of Sweden, but are most profound in Västergötland, Skåne, Öland, Gotland, Närke and the Jämtland region. The shale was earlier used as part of aeriated concrete.

In other bedrocks, such as granitoids, that originates from a melted or partially melted magma, an enrichment of radioactive elements is common (Jelinek & Eliasson, 2015). For example, K-feldspar, that constitutes part of micas is often enriched in a late stage crystallization of a magma. Generally, accessory minerals, such as monazite, zircon, apatite or allanite, containing the radioactive isotopes uranium and thorium, also accompany the late stage crystallization phases (Klein & Hurlbut, 1993). The relation between the mineral composition in Swedish bedrocks and the minerals containing the natural radioactive elements is well described and summarized by Jelinek & Eliasson (2015). An overview of Swedish bedrock and its gamma radiation levels, as well as contribution to the release of radon gas may also be found in Møre (1985), Petterson et al. (1982), Mjönes et al. (1984) and Åkerblom & Clavensjö (2004, 2007).

For concrete the corner stone was earlier natural aggregates (glaciofluvial sediment) but are more commonly today crushed bedrock (aggregates) in combination with cement and water. All three constituents, that are the basis for concrete, can accordingly contain substantial amounts of the radioactive isotopes 40K, 232-Th and 238U.
1.2 Aim of the Thesis

The aim of the Thesis is to gain an increased understanding of natural radioactivity within the field of construction and building materials. For radiation protection, different quantities are used. The Thesis aims to clarify the use of the different quantities and the links that could be established between calculated models used for protection purposes and the measurement techniques applied on a daily basis. The Thesis approaches these issues by using different measurement techniques for detection of gamma radiation within concrete. The Thesis has also focused on ionizing radiation from the uranium chain ($^{238}\text{U}$) emitting radon gas ($^{222}\text{Rn}$). The relation of relative humidity (RH) and its influence on the radon gas exhalation from concrete blocks also constitute a major part of the Thesis. The regulations, as of today state, not only to the effective dose given by the gamma radiation, set as 1 mSv per year as the maximum reference value from construction materials (EC, 2014), but also the radon gas limitations as set by the National Board of Housing, Building and Planning (2006) at 200 Bq/m$^3$ give an input to some of the identified research questions during this project.

1.3 Research questions

RQ 1. Is there a good relationship between different measurement techniques of ionizing radiation from the natural radioisotopes, $^{40}\text{K}$, $^{226}\text{Ra}$ and $^{232}\text{Th}$?

The research questions aims to clarify correlations between (i) a gamma spectrometric laboratory equipment, (ii) a hand held portable gamma radiation equipment and (iii) geochemical analysis.

RQ 2. Is it possible to quantify the activity concentration (Bq/kg) of crushed aggregates (constituent of concrete) and the concrete composed of the very same aggregate? and secondly, what difference in annual dose rate, expressed in mSv per year could be expected using only aggregates and when the aggregates constitute part of the concrete?

The research questions highlight the calculation of the final gamma radiation dose within a building (room) by use of theoretical models presented in papers from the EU-commission and other sources.

RQ 3. Is radon exhalation (Bq/m$^2$h) affected by the relative humidity (RH) of the concrete?

The research question is vital for a good understanding of the exhalation rate within concrete due to its relative humidity and the possible prediction of the radon gas rate (exhalation rate) thereof.

RQ 4. Do different water-cement ratios (w/c ratios) influence this possible correlation between RH and radon gas exhalation?

In the project two different water-cement ratios have been used for casting of concrete and evaluation.

RQ 5. How does the radon gas exhalation rate influence the final rate of radon gas within a predefined room?

A calculation of the exhalation rate of radon gas of the cast concrete specimens making use of the limit values of air circulation rate within a room as defined by the by the National Board of Housing, Building and Planning has been assessed.

1.4 Limitations

The assessment of investigating different measurement techniques for gamma radiation is limited to ten different aggregate materials, due to the time frame and budget of the project. In view of sample preparation and uncertainty factors, there is also a limitation due to handling of concrete specimens. The full size of the aggregate mixtures and concrete castings could not be assessed, and hence smaller portions are viewed as representative of larger samples.
The evaluation of the radon gas exhalation rate initially posed a problem. This was due to limited measurements of each concrete sample caused by limited access to the radon gas laboratory for assessments. This was a conflict related to loads of coinciding commission work. Hence, only a few measurements could be performed for some samples.

Another limitation within the project is the in part shortcoming to sufficiently explain the different theoretical radiological models used within the project. As an example, the project does not fully cover the derivation of the isotropic source point model including the process of attenuation and build up according to the Berger model. The limitation is due to some constants used within formulas, which are derived through either tables in reports (NIST, ICRU) or complex calculations (such as “Attenuation and Build up” or “Monte Carlo Simulations”). This would give rise to further continuous questions within radiation protection and the different quantities used. Furthermore, the operational and measurable quantities could only be addressed with a glance at the surface. The complexity to evaluate different deterministic and stochastic effects to the human body pose a difficulty for many experts as well to agree on a mutual agenda concerning regulatory outlines of the different risks with particles, photons, alpha-particles, neutrons and muons. For a deeper knowledge of the different quantities used the readers are primarily advised to read ICRP publication 103 (ICRP, 2007) or Isaksson (2011a, b).

The equipment used within the project, such as ATMOS 33 delivered by Gammadata AB and the hand-held spectrometer delivered by Radiation Solutions Incorporated and measurements of the relative humidity (Vaisala Oy) would indeed need a deeper technical investigation. However, this causes the scope to be shifted into measurement techniques, which has not been intended as the focus of the Thesis. Different measurement techniques should also be evaluated more thoroughly and the author sees this as vital, since many different techniques, such as passive and active techniques could give rise to arguable correlations (Al-Jarallah et al., 2001) and subsequently certain mistrust in the equipment.

1.5 Content of the Thesis

The content of the Thesis embraces some aspects of radioactivity of natural isotopes of current interest, whilst the European Union is trying to consolidate and set some harmonized standards for the use of specific techniques within the field of radiation for construction purposes.

Hence, it follows that the Thesis entails three papers with a content describing a comparative approach of different measurement techniques and some errors, worth highlighting, that could easily sum up to incorrect assessments of the final results (Paper I). Paper II focuses on comparison of the European assessment tool, the I-index to evaluate the potential risk of elevated doses in conjunction with several other assessment tools, such as the radium index, alpha-index and some common international indices such as the H_{eq}-index to evaluate the gamma radiation in dwellings, Paper III makes use of the I-index and compare it with an assessment tool, recently developed by the Hoffman (2014), as to more precisely show the differences between the I-index as a conservative tool and a more accurate tool to evaluate the final dose to human beings using a specified concrete recipe. In short the papers’ content are presented below.

**Paper I** - A comparative study of different techniques for the evaluation of gamma radiation

**Paper II** - Comparative study of different indices of natural radioactivity related to gamma radiation and radon gas contributions.

**Paper III** - The current EU suggested assessment tool, the activity concentration index, I, used for the evaluation of building materials and a dose model, which has been used for the evaluation of some single Swedish crushed aggregates and its final concrete specimens.

The Thesis has a primary focus to the gamma ray decay of (natural) radiation and secondly the nature of radon gas exhalation of the concrete specimens produced within the project. This is partly due to the focus of the current EU-legislation, where the European guidelines (CPR, 2011, EC, 2014) of
thresholds of gamma ray radiation are set to half of the current recommended value of the Nordic countries (The Radiation Protection Authorities in Denmark, Finland, Iceland, Norway and Sweden (2000), which poses a dilemma for some contractors and concrete producers. The author also believes it is vital to understand the links between the units and proposed dose models, which are used as guidelines to some reference values, that should not be exceeded (EC, 2014).

The initial Chapters (1 and 2) of the Thesis entail the fundamentals of ionization, decay of elements (e.g. $^{238}$U and $^{232}$Th) and the resulting products with their different characteristics. The Chapter also includes the impact of different energies, materials different attenuation and the resulting effect shown by some basic calculations that can be made to understand the impact of different energies. A more explanatory section is introduced in Chapter 3 as to understand some of the different quantities and units used within radiation protection. Within this Chapter also some vital links between absorbed dose, ambient dose, equivalent dose and effective dose are explained. In the following section (Chapter 4) a description of the methodologies used and specifically the technique and use of the portable hand-held spectrometer are explained. The essential characteristics of the instrument are further explained, its working principles, calibration thereof and units used as to convert between Röntgen (R), absorbed Dose (D), Ambient Dose, $H^*(10)$. The chapter also encompasses the conversion of elemental concentrations of $^{40}$K, $^{226}$Ra and $^{232}$Th and how these are converted to activity in Bq/kg and the total dose (ambient dose rate) as presented in the results.

In Chapter 5 an explanatory part includes the general characteristics of radon gas and its behaviour in concrete (and partly soil) and the instrumentation used to measure radon gas. Furthermore, the instruments principles are described and its functionality. An explanation of the calculation principles of the exhalation rate of radon gas in air is also included. Moreover, a small section describing the calculation principles of radon gas in a room, including examples finalizes this chapter in conjunction with the different set ups used within this Thesis.

In Chapters 6 and 7 the results are presented in relation to gamma radiation and radon gas, respectively. The results are followed with a discussion (Chapter 8) of the results, comparative investigation and uncertainties. A final summary (conclusions) of the results are presented in Chapter 9 accompanied by proposals for future research (Chapter 10). The references are found in Chapter 11 followed by the presented papers reprinted as Appendices to the Thesis.
2 Ionizing radiation

2.1 Fundamentals

Ionizing radiation is radiation that carries enough energy to free electrons from atoms or molecules, thereby creating ions in the air, thus ionization occur (Isaksson, 2011a).

The basis of ionizing radiation is due to the fact that heavy atoms are unstable. Thus, the unstable atomic nuclei decay in order to find a more stable form. This phenomenon generating different chains of decay is a consequence of a heavy atom searching for stability while sending out radiation as α-particles or β-particles, sometimes including an excess of energy, named gamma radiation (Isaksson, 2011a). The gamma rays have sufficient energy to cause ionization of matter and therefore can cause damage to living cells (Stalter & Howarth, 2012). In Figure 2.1 the decay of $^{232}$Th is represented to show the complex degradation of different isotopes. Within the circles presented below the total number of protons and neutrons (upper left) is given as well as the atomic number (number of protons in the nucleus) together with the chemical symbol in bold text. Below the elements the half-lives of each element are given. The decays are indicated by arrows, denoted α (alpha-emission) or β (β-emission). Gamma emission (γ) often follows β-decay. However, this is not depicted in Figure 2.1. α-emission causes the element to emit two neutrons and two protons, meaning the mass number decreases by four, while the atomic number (number of protons) decreases by two. In the decay chain, negative β-emission (emission of an electron and anti-neutrino) also converts a neutron into a proton, hence leaving the mass number intact, while the atomic number increases by one, as depicted in Figure 2.1.

Figure 2.1 The $^{232}$Th decay chain illustrating the change of its elements during alpha- and beta emission. Within the figure the gamma emissions are not presented. Based on the Thorium SVG image by contributor BatesIsBack.

$^{232}$Th itself is not measured by gamma spectrometry techniques, but its daughter isotopes $^{228}$Ac or $^{208}$Tl are often used (Klemola et al., 2010). As a result, this requires equilibrium between $^{232}$Th and its daughter isotopes, in order to make correct calculations. This scenario is exactly the same for $^{238}$U/$^{226}$Ra, where measurements are performed on its daughter isotope $^{214}$Pb. The only radionuclide,
which is measured directly using gamma spectrometry technique, is $^{40}$K. Using a portable spectrometer, most often, $^{214}$Bi is used instead of $^{214}$Pb, which is normally used in a laboratory. 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 has to be 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.

For both $^{238}$U ($^{226}$Ra) and $^{232}$Th the initiation of the breakdown of the nucleus and consequently the decay chain initiation starts with alpha decay (Cember & Johnson, 2009). As the decay progresses also beta emissions- and gamma radiation are followed changing the parent atom to different phases. As an example, the $^{238}$U atom decays to various alkali-metal ions, radon gas and its daughter isotopes ($^{218}$Po, $^{214}$Po), and finally reaches a the stable lead isotope $^{206}$Pb (Isaksson, 2011a; Cember & Johnson, 2009). In Figure 2.2 the decay chains of $^{238}$U and $^{232}$Th are exemplified. The release of alpha-, beta- and gamma energies at different energy levels are also highlighted.

![Radioactive Decay in Thorium and Uranium Series](image)

**Figure 2.2** The $^{232}$Th and $^{238}$U decay chains illustrating their emissions of alpha, beta and gamma radiation, as well as the release of gamma energies and some half-lives of nuclides. Image from World Nuclear Association.

### 2.1.1 Alpha emission

The alpha particle is a double-charged particle, quite slow in velocity and hence quite easily stopped. It is identical to a helium-nucleus, consisting of two neutrons and two protons. The double positive charge of the ion attracts other electrons from their parent atoms. Hence, when alpha-particles are emitted through a material, some electrons will be attracted and depart from their outer parent atom shells and consequently ions will be produced in the material. In other words ionizations occur (Isaksson, 2011a).

An alpha-particle is a large particle compared to beta-particles and especially photons (gamma rays), which are considered as only energy quanta or electromagnetic radiation, constituting no weight. An alpha-particle weighs 8000 times more than a beta-particle, which is basically an electron (Isaksson,
Table 2.1 depicts some of the characteristics of different energies, which results due to the ongoing decay of natural radionuclides.

Table 2.1. Some characteristics of α, β particles and γ energies.

<table>
<thead>
<tr>
<th>Particle</th>
<th>Constituents</th>
<th>Charge</th>
<th>Comparative weight</th>
<th>Energy (keV)</th>
<th>Wavelength (m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>α</td>
<td>2 protons + 2 neutrons</td>
<td>+2</td>
<td>8000</td>
<td>~ 4 900</td>
<td></td>
</tr>
<tr>
<td>β</td>
<td>1 electron</td>
<td>-1</td>
<td>1</td>
<td>800-1400*</td>
<td></td>
</tr>
<tr>
<td>γ</td>
<td>Energy</td>
<td>-</td>
<td>-</td>
<td>&gt; 100</td>
<td>&lt;10^{-12}</td>
</tr>
</tbody>
</table>

*Most frequent energy-interval of a beta-particle where the maximal value of its energy equals approximately 2.5 MeV.

2.1.2 Beta emission

Beta-particles are negatively charged particles, with a very high speed (close too speed of light). The negative charge of the particle will deflect other electrons in their parent atoms. Due to speed and closeness to other electrons, it could deflect electrons out of their parent atom shells, and as such, ionization occurs. Beta-particles can also have a positive charge (β⁺). This occurs if a nucleus has an excess of protons. The reaction involves a conversion of a proton to a neutron (positron) and a neutrino (Isaksson, 2011a). Beta particles can at higher energy ranges (> 1 MeV) penetrate a humans skin (Turner, 2007)

2.1.3 Gamma ray emission (gamma rays)

Gamma ray emission occurs as a consequence of alpha- or beta decay. (Isaksson, 2011a). Gamma radiation could also be described as electromagnetic radiation with very high frequency, i.e. with energies above 100 keV and wavelengths less than 10 pm (10^{-12} meter), which is less than the diameter of an atom (Isaksson, 2011a). Gamma radiation is thus a form of electromagnetic radiation, such as x-rays and light. The difference between x-rays and gamma rays are dedicated to the origin of the ionising radiation source. For gamma rays, the origin is at the nucleus as a consequence of decay, whereas for x-rays the origin is related to the electronic shells (M, L, K) of the atom (Cember & Johnson, 2009).

The ionizations of material by gamma rays occur through several processes. Two of importance are (i) compton scattering and (ii) the photoelectric effect. These are the most prominent ones in the field of measurements for natural radiation (40-3000 keV). At higher energies, yet another process, called pair production, increases in importance (Turner, 2012).

Compton scattering occurs when a high energy photon transmits some of its energy to another electron and deflects it (pushes it out of the parent atom shell). The remaining energy of the photon continues in another direction (scattered) through the material.

The photoelectric effect is slightly different compared to compton scattering. Here the incident photon rams into an electron, transfers its energy into the electron, which is ejected out of its parent shell (ionization) and a photoelectron is the final product, which continues its transfer in the material. At the end the loss of energy to other electrons will result in absorption of the reminder of the energy by a random parent atom (Isaksson, 2011a).

Pair production occurs when a photon is converted to one positron and one electron. (Isaksson, 2011a).
2.2. Material characteristics and energy transfer

2.2.1 Density

The density of a material affects the absorption of ionizing radiation. The denser the material, the more closely spaced are the atoms, generating a large impact for the incident gamma energy to be absorbed into the material (Turner, 2007).

2.2.2 Atomic number

The absorption capabilities of gamma rays are largely proportional to the atomic number and the density. As such a low atomic number as for oxygen, almost repeal no gamma rays. As for lead with an atomic number of 82, this factor is on the contrary. It’s an efficient absorber of gamma rays.

2.2.3 Attenuation of gamma rays

A common way to describe the effectiveness of gamma ray reduction is through “half-layer value” or attenuation coefficient of different materials. A high attenuation coefficient is consequently characterizing a strong absorber of gamma rays.

Table 2.2 presents a list of different materials with different attenuation coefficients at energies 100, 200 and 500 keV, as well as the natural radioactive isotopes and their attenuation coefficients in concrete at their most prominent energies (Turner, 2012) and data from Hubbel & Seltzer (2004) - NSTIR 5632.

Table 2.2 A few absorbers/materials and different attenuation coefficients due to different energy intervals and radioisotopes.

<table>
<thead>
<tr>
<th>Absorber (material)</th>
<th>Energy (keV)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>100</td>
</tr>
<tr>
<td>Concrete – ^{238}U</td>
<td></td>
</tr>
<tr>
<td>Concrete – ^{232}Th</td>
<td></td>
</tr>
<tr>
<td>Concrete - ^{40}K</td>
<td></td>
</tr>
<tr>
<td>Air</td>
<td>0.000195</td>
</tr>
<tr>
<td>Water</td>
<td>0.167</td>
</tr>
<tr>
<td>Carbon</td>
<td>0.335</td>
</tr>
<tr>
<td>Aluminium</td>
<td>0.435</td>
</tr>
<tr>
<td>Iron</td>
<td>2.72</td>
</tr>
<tr>
<td>Copper</td>
<td>3.8</td>
</tr>
<tr>
<td>Lead</td>
<td>59.7</td>
</tr>
</tbody>
</table>

2.2.4 Attenuation of alpha emissions

As earlier mentioned, reduction (attenuation) of alpha particles as to reduce the rate of radon gas exhalation can be controlled by different variables. One is to paint the walls or put up wall paper. This significantly reduces (~30-50%) the radon gas release (Petterson et al., 1982). However, when evaluated as part of the initial concrete product, the radon gas content is viewed in the light of the
material characteristics. A second approach to regulate the rate of radon gas exhalation emitted from building materials inside the room is through circulation of air, meaning ventilation of outside air into the room.

2.2.5 Intensity and calculation of reduction of energy

Knowing the attenuation coefficient of a material a simple calculation of the total energy reduction could be assessed (Turner, 2012).

For example, how much aluminum is required to reduce the intensity of a gamma ray to 10 per cent of its initial energy knowing that the photon energy is 200 keV ?

From Table 2.2 in subsection 2.2.3 we find that the attenuation coefficient of aluminum for the energy 200 keV is 0.324. The following applies

1. The linear attenuation coefficient is denoted as, \( \mu \)
2. The thickness (in cm’s) required, denoted as, \( x \)
3. The initial energy of an energy gamma ray beam is described as \( I(0) \).

\[
Ix = \frac{I(0)}{10}
\]

\[
Ix = I(0) \exp(-ux)
\]

\[
I(0) \frac{1}{10} = I(0) \exp(-ux)
\]

\[
\frac{1}{10} = \exp(-0.324x)
\]

\[-\ln 10 = -0.324x
\]

\[x = \ln 10/0.324
\]

\[x = 7.1 \text{ cm}
\]

- If we were to use lead instead for the same photon energy, the required thickness of lead would be 0.23 cm.
- However at an energy level of 500 keV, the required thickness increases to 1.4 cm in order to reduce the initial radiation to 10 %.
- Thus the energy increase (in keV) has a major input to the attenuation coefficient of the material. And for the natural isotopes as part of concrete, using the attenuation coefficients presented in Table 2.2, only ten percent of the emitted photons would remain for \(^{238}\text{U}\) after 14 cm of concrete, and for \(^{40}\text{K}\) after 19 cm and for \(^{232}\text{Th}\) after 25 cm, respectively.
3 Gamma radiation and radon gas

3.1 Gamma radiation and dose quantities (external radiation)

3.1.1 General

The different quantities used for quantifying radiation are numerous and not easily kept apart. In Figure 3.1, a modified flow chart (Isaksson, 2011b) are presented in order to clarify the relationships between physical, operational and monitoring as well as protection quantities. A necessity within this Thesis has been to shorten some relations. Explaining all different relations and different ways of calculating different doses are plentiful. For a thorough understanding the reader is advised to review and study the Annals of the ICRP (2007).

One way to describe some of the correlations between different quantities is to describe the links through a flow chart (Figure 3.1). The most important quantities and units are also summarized in Table 3.1.

Figure 3.1 Different quantities and their linkage described by a simplified flow chart. Modified from Isaksson (2011b), [http://www.wikiwand.com/en/ionizing_radiation](http://www.wikiwand.com/en/ionizing_radiation). Red marked rectangles highlight one of the procedures used during this Thesis, by using a portable hand-held spectrometer to measure, calculate and compare the ambient dose equivalent of an object to the effective dose.
Table 3.1 Quantities, units and definitions for ionizing radioactivity.

<table>
<thead>
<tr>
<th>Quantity</th>
<th>Unit (full name)</th>
<th>Unit (abbreviation)</th>
<th>Definition/ short explanation</th>
</tr>
</thead>
<tbody>
<tr>
<td>Radioactivity</td>
<td>Becquerel</td>
<td>Bq</td>
<td>One Bq is defined as the activity of a quantity of radioactive material in which one nucleus decays per second.</td>
</tr>
<tr>
<td>Absorbed dose (D)</td>
<td>Gray (Gy)</td>
<td>1 Gy = 1 J/kg</td>
<td>Represents the energy absorbed by the human tissue. Basic physical dose quantity. Mean energy imparted to matter/ mean mass. Stochastic quantity – measurable.</td>
</tr>
<tr>
<td>Exposure (X)</td>
<td>Roentgen</td>
<td>1 R = 2.58 \times 10^{-4} C/kg</td>
<td>Measure of the ionization of uncharged particles produced in air by X-rays or gamma radiation. Link between the units Roentgen (R), absorbed dose (D) and ambient dose rate, H*(10)</td>
</tr>
<tr>
<td>Ambient equivalent dose H*(10)</td>
<td>Sievert (Sv)</td>
<td>1 Sv = 1 J/kg</td>
<td>Estimate of Equivalent dose/ effective dose for area monitoring, measured opposite the radius of the incident photons within a sphere at 10 mm’s depth.</td>
</tr>
<tr>
<td>Equivalent dose (H_T)</td>
<td>Sievert (Sv)</td>
<td>1 Sv = 1 J/kg</td>
<td>The Sievert represents the equivalent biological effect of the deposit of 1 J of ionizing radiation energy in a kilogram of human tissue. The dose equivalent encompasses the radiation weighting factor, W_R, which depends on the type of radiation (D_T,R) – absorbed dose. Not a measurable quantity.</td>
</tr>
<tr>
<td>Effective dose</td>
<td>Sievert (Sv)</td>
<td>1 Sv = 1 J/kg</td>
<td>The effective dose is the summation of all different equivalent doses by a factor, W_T, as defined by the ICRP, 2007. Assuming full body irradiation for gamma rays (photons) the Equivalent dose and Effective dose could be considered equivalent.</td>
</tr>
<tr>
<td>I-index</td>
<td>-</td>
<td>Dimensionless</td>
<td>Conservative estimate of the maximum “effective dose” under circumstances, where a given “point-dose” is calculated within a room (Markkanen, 1995). Specific conditions presented in RP 112 (EC, 1999).</td>
</tr>
</tbody>
</table>

3.1.2 Physical quantities

The physical quantities are sometimes described by means of fluence rate ($\phi$) and depend on the incoming irradiation (geometry). The other physical quantities are Kerma $K$ and absorbed dose $D$. The Swedish Authority of Radiation and Safety (SSM) earlier used Kerma (Kinetic energy release per unit mass) as a mean to calculate the absorbed dose (Andersson et al., 2007) taking the bremsstrahlung into consideration. The following relation is valid:

$$D = K (1 - g)$$  \hspace{1cm} (3.1)

Where $g$ denotes the loss of energy in bremsstrahlung (Isaksson, 2011a). Since the loss of energy due to bremsstrahlung often is restricted the kerma rate ($K$) could in air be approximated as equal to the absorbed dose. The ICRP (2007) recommends the assessment of absorbed dose as the fundamental physical quantity to assess effective dose. The absorbed dose is basically a measure of the energy (mean) imparted in a small volume due to irradiation (ICRU 1998). It is defined as:

$$D = \frac{d\varepsilon}{dm}, \text{ where } \varepsilon \text{ is the energy and } m = \text{ the mass in kg}$$  \hspace{1cm} (3.2)
Several different irradiation situations are applicable (ICRP, 1996), but the most commonly used models to describe the irradiation are rotational, ROT (rotational) and ISO (isotropic) according to The Radiation Protection Authorities in Denmark, Finland, Iceland, Norway and Sweden (2000), Isaksson (2011b) and ICRP (2007). A rotational situation could be approximated by area monitoring, using field equipment (ICRP, 1996). An isotropic situation could be simulated as a dose point (Chabot 2007), which is described by Markkanen (1995), when calculating the dose within a room where the irradiation comes from all directions.

The physical quantities can be used to calculate risk related quantities (protection quantities) by use of weighting factors or conversion coefficients as could be found in different reports, such as the ICRP-report 74 (ICRP, 1996) and later additions (ICRP, 2006, 2009).

3.1.3 The ICRU-sphere

In order to calculate different risks a phantom, representing the human body is defined, the so called ICRU-sphere. The ICRU-sphere is a 30 cm diameter tissue equivalent sphere consisting of a material with a density of 1000 kg/m³ and a mass composition of 76.2 % oxygen, 11.1 % carbon, 10.1 % hydrogen and 2.6 % nitrogen. This material is called ICRU-tissue.

3.1.3 Monitoring and operational quantities (measurable)

The monitoring quantities (see Figure 3.1) are those that are used for measurements. Hence, to be valuable the instruments intended for use need to be calibrated accordingly. At least four different ways to calibrate your equipment are described by IAEA (1989).

In Sweden, a national specific site for calibration of geophysical instruments is available. At Borlänge airport, large calibration pads (concrete bricks in a circle) are used with well-defined levels of natural radiation within the material (Figure 3.2).

Hence the detectors as part of the portable instruments can be calibrated and adjusted in relation to well defined levels.

Figure 3.2. Calibration site at the Borlänge airport. The calibration pads used to calibrate instruments that measures the content of the natural radionuclides of $^{40}$K, $^{226}$Ra, $^{232}$Th. The pads are shown with asphalt in between. In the foreground an ongoing calibration of the RS 125 BGO spectrometer is seen. The figure is presented with the permission of Leif Löfberg.
The operational quantity, exposure \((X)\), although not frequently used today, are essential to some of the links between absorbed dose rate and ambient dose rate \(H^\ast(10)\) as well as equivalent dose rate \((H^\ast)\). Given that the energy, or exposition of \(2.58 \times 10^{-4} \text{ C/Kg} = 1 \text{ R}\), it also follows that this relation gives the condition that \(1 \text{ R} = 8.69 \times 10^{-2} \text{ Gy in absorbed dose in free air}\). Exposure is defined as the sum of electric charges \(dQ\) of one sign of ions liberated indirectly by photons of gamma- or x ray radiation in air divided by the mass of air \(dM\) (IAEA, 2003). Accordingly, this is a fundamental relation often used by hand-held spectrometers in order to establish relationships between different units.

The operational quantities are defined by the relation of a specific dose (absorbed dose = \(D\)) at a given distance within the body \(H^\ast(x)\). For a normal situation of irradiation to the skin a given distance of 10 mm is recommended by the ICRP (2007). Thus, the dose quantity ambient equivalent dose is written as \(H^\ast(10)\), representing a point 10 mm’s into the human body. Moreover, the absorbed dose rate \((D)\) could be converted to the risk related quantity (protection quantity) equivalent dose rate as,

\[
H = D \times W_R \quad \text{(see also Table 3.1)}
\]

\[
(3.3)
\]

Where \(W_R\) is quality factor\(^1\) depending on the kind of radiation that is emitted (Section 3.1.4), relating to the damaging of the biological tissue (relative biological effectiveness) and \(D\) represents the absorbed dose at that specific point in the human body (ICRP, 2007). For photons or \(\beta\)-particles a \(W_R\)-value of 1 is assigned. However for alpha-particles (radon and its progenies) a quality factor of 20 is applicable (ICRP 2007).

For environmental investigations, as within this study, the Ambient dose equivalent, \(H^\ast(10)\) is often of interest, since it could be related to the effective dose (Isaksson, 2011b, ICRP, 1996). The ambient dose equivalent \((H*d)\) or often represented as \(H^\ast10\), is the dose equivalent that would be produced by the corresponding expanded and aligned field in the ICRU sphere at a depth \(d\), on the radius opposing the direction of the aligned field (IAEA; 2000).

![Figure 3.3 Radiation geometry of the ICRU sphere to determine ambient equivalent dose, \(H^\ast(d)\) in an expanded and aligned radiation field. The radius vector always opposes the direction of the radiation field.](image)

The measurements conducted with the portable hand held spectrometer use the ambient dose equivalent rate as to evaluate the effective dose conservatively. The absorption of photons (gamma rays) in relation to a given geometry varies quite strongly in the interval 10-3 000 keV. Hence a relation between geometries (e.g. ISO/ROT – geometry) is often given at a specific energy. For the risk related quantity, \(E\), effective dose in relation to \(H^\ast(10)\) a common approach is to describe it in relation to the energy of 1 MeV (The Radiation Protection Authorities in Denmark, Finland, Iceland, Norway and Sweden, 2000) as,

\[
E = H^\ast(10) \times 0.6 \quad \text{(ISO- geometry)}
\]

\[
(3.4)
\]

\(^1\)\(W_R\) was earlier defined as "\(Q\). For photons and alpha-particles the radiation factors defined by the "\(Q\)"-value is equal to the \(W_R\) factor (ICRP, 2007).
The relations between different geometries for photons (gamma rays) are also found in ICRP (1996) and shows that the ambient dose rate could never over-estimate the effective dose.

A more precise relationship between operational quantities and risk related quantities (that cannot be measured) could be assessed by using the operational quantity, personal dose equivalent, \( H_p(d) \), often described as \( H_p(10) \). \( H_p(10) \) is considered comparable to the effective dose by ICRP (2007). The personal dose equivalent \( H_p(d) \) should be used for soft tissue and for humans under investigation (Table 3.1). It can be measured by use of dosimeters that are worn in direct contact with your skin (Erlandsson & Isaksson, 2006). A common type is thermo luminescence dosimeters (TLD) consisting of lithiumflouride (small chips). Its correlation to the risk related quantity, the effective dose could be found by knowing the air kerma and reading of the instrument.

The overall radiation (terrestrial radiation) including the cosmic radiation is often measured by means of portable equipment (hand-held). Common instruments are intensimeters (GM-counters) or spectral high resolution scintillation analyzers, such as the instrument used by CBI (using a Bismuth Germinate oxide crystal for enhanced resolution).
3.1.4 Protection quantities

These are quantities that cannot directly be measured. However they can be calculated through investigation and by use of numerical links or dose conversion factors (ICRP 1996, 2012). The equivalent dose \( (H_T) \) makes use of the calculated absorbed dose and takes all different parts of the body into consideration. The equivalent dose is specified for calculation of separate parts of the body, e.g. the gonads. By use of a weighting factor \( (W_R) \), depending on radiation type (photons, neutrons or electrons), consideration is taken to this specific part/organ. Secondly, the equivalent dose takes into consideration the different values \( (D_{T,R}) \) assigned to different organs of the body, due to their sensitivity to radiation. The equivalent dose to a specific organ or tissue \( T \) is defined as

\[
H_T = \sum_R W_R D_{T,R} \tag{3.5}
\]

The above radiation type, \( R \), \( W_R \) and \( D_{T,R} \) values to each specific organ or parts of organs could all be found in different tables of ICRP (2007), section B. Different organs also has its weighting factor \( (W_T) \) defined in ICRP (2007). If the whole body is exerted to homogenous irradiation, \( W_T \) could be assigned as equal to one (meaning all organs are irradiated). Under those conditions the equivalent dose in magnitude can be considered as equal to the effective dose \( (E) \) according to

\[
E = \sum_T W_T H_T \tag{3.6}
\]

Thus, if we are to assume that the effective dose \( (E) \) is due to only photons and full body irradiation occurs we could make use of a formula presented by IAEA (2003) as how to calculate the effective dose generated by building materials using the numerical links calculated by Markkanen (1995) according to,

\[
E = D' \times t \times 0.7 \times 10^{-6} \tag{3.7}
\]

where, \( D' \) = absorbed dose rate in air (nGy/h). The absorbed dose rate in air could be calculated by knowledge of each radioisotopes specific activity (Bq/kg) \( \times \) conversion coefficients, \( t \) = hours spent indoors, 0.7 = conversion coefficient between Sv/Gy for human organs (UNSCEAR, 1988). The result is reported in mSv.

3.1.5 Relation between Effective dose \( (E) \) and I-index

In the European Commission report (EC, 1996) dealing with natural radioactivity in building materials some of these numerical links to establish a relation between “measurable” and “protection” quantities have been calculated using an isotropic source point including the attenuation and build up factor by the Berger model. The dose \( (D) \) at a specific point, taking into account the shielding of a material and scattered secondary photons adding to the dose, defined as buildup \( (B) \) and using iso-geometry the dose can be calculated as:

\[
D = kSE \frac{\mu_{en}}{\rho} Be^{-\mu T} \div 4\pi r^2 \tag{3.8}
\]

Where \( E \) is the photon energy, MeV, \( \mu_{en} / \rho \) is the mass energy absorption coefficient for the material at the dose point, cm\(^2\)/g, \( k \) is a collective constant to convert energy fluence rate to absorbed dose rate (gray/hour = \( 5.76 \times 10^{-7} \)), \( T \) is for thickness of shielding source and \( u \) = linear attenuation coefficient for energy of interest in the shielding material and \( S = \) source emitting gamma rays/second and \( r \) = distance from source \( (m) \). \( B \) stands for the buildup factor and is defined as,

\[
B = 1 + a\mu T e^{b\mu T} \tag{3.9}
\]

\(^2\) Conversion coefficients have been calculated for given geometries in a room, e.g. Markkanen (1995) and RP 112 (EC, 1999) presented specific dose rates (nominal doses) in the unit nGy/h per Bq/kg of \(^{40}\)K, \(^{226}\)Ra and \(^{232}\)Th.
where \(a\) and \(b\) are constants for a given energy and shield material.

The formula used to describe the dose contribution within a room rests on this principle but are described in detail in the appendix of STUK STO-32 (Markkanen, 1995).

The calculations and the results of some numerical links (specific dose rate in air (nominal dose) of the radioisotopes, \(^{40}\text{K}, \quad ^{226}\text{Ra} \quad \text{and} \quad ^{232}\text{Th, in relation to thickness of the shielding material) are further described in report} \quad 112 \quad (EC, 1999) \quad \text{and serve as the basis for calculation of the effective dose to the human body within a given room volume (4x5x2.8 m). It should be noted, that these numerical links, as D’ in formula 3.7 above (subsection 3.1.4) have been calculated by different authors (Manic, 2012, Markkanen, 1996, Beretka \\& Mathews, 1987, Mustonen, 1985, Stranden, 1977) and could differ considerably (±30 %).}

However, the European Commission (EC, 2014) has set a recommendation that building materials are not to exceed 1mSv per year in effective dose. This approach considers geometrical arrangement of a human situated within a room of \(3 \times 4 \times 2.5\) m spending 80 % of the time within that specific room (EC, 2014). Thus, due to the change of the dimensions of the room, the effective dose to a human body, will in the revised Draft TR 00351020 be shown to be somewhat higher than previously presented (EC; 1999, Markkanen, 1995). However, the difference is only a few % compared to earlier calculations.

Consequently, in order to find a tool to establish a comparable link between the effective dose (isogeoetry) exerted to the human body within this predefined volume, Markkanen introduced the dimensionless I-index as a way to roughly compare a value of ≤ 1mSv in effective dose per year to the value of 1 in I-Index given the above conditions. The I-index is supposed to serve as a screening tool and a simplified instrument to use for the public, contractors and regulators, with the purpose to evaluate their building material used within a specified constructional work. However, the relation between the I-index and effective dose per year is only valid under the conditions of a predefined room, using a concrete of 2350 kg/m\(^3\), where the thickness of the walls, roof and floor is ≤200 mm. The use of I-index is adopted by the EC’s Basics Safety Standards (2014), where Annex VIII specifically describes the I-index formula and its limitations.

Consequently, the protection quantities, serve as tool for regulators, contractors and all parts within the construction chain as to ensure a good environmental conditions, within a building as well as within the urban areas.

Even though not dealt with in detail within the scope of the Thesis, calibration coefficients relating to dosimetry could be found in report ICRP 74 (ICRP, 1996), where numerous different geometries are calculated and graphically describes the influence of different geometries (AP, PA, LAT, ROT, ISO) in relation to the effective dose and kerma rate as a function of the energy of the photons. Explanations of the different quantities and units could further be assessed in report ICRU 60 (ICRU, 1998). More conversion factors within radiation protection are to be found in different reports by the ICRP (ICRP, 1996, 2006, 2009, 2012). In Figure 3.4 is depicted the difference between measuring the ambient equivalent dose rate (\(H*10\)) compared to effective dose rate in a isometric (ISO) radiation field: It also relates to Equation 3.4, since the ratio of \((E/Ka)/(H*10/Ka)\) equals approximately 0.6 (iso-geometry).
3.2 Radon gas and dose quantities (internal radiation)

3.2.1 General

The ionization of human cells due to the decay of radon gas and its progenies through alpha particle emission is caused by the decay of $^{222}\text{Rn}$, as well as the daughter isotopes $^{218}\text{Po}$ and $^{214}\text{Po}$, which all emits alpha particles. The largest risk with radon gas is as such the inhalation by air where different organs are exposed to different degrees of radiation. The intake of food and drinks also relates to the same subject. But, related risks are most fiercely, when alpha decay occurs. As such the whole uranium- and thorium decay chain as alpha-emitters have to be considered (Figure 2.2).

3.2.2 Monitoring and operational quantities

Different techniques are applied to measure the content of radon gas. The techniques may be classified as active or passive (Isaksson, 2011a, Knoll, 2010). Active detectors require electrical power to measure the diffusion of radon gas. Passive techniques do not require any power source.

Some active techniques are:

1. **Solid surface barrier detectors**, making use of alpha spectrometry. Filters are used to separate each radon progeny in order to enhance reliable calculations.

2. **Scintillation cells** are enclosed cylinders coated with scintillation material, such as silver activated zinc sulphide powder. Interaction of alpha particles with the scintillation material causes production of light photons, which are detected and amplified by a photomultiplier and converted to an electrical signal. Each progeny could be distinguished by complementary counting techniques and as such the methodology is considered to give high resolution and hence good control of uncertainties.
3. **Ionization chamber** has a central collecting anode within a metal cylinder. The alpha particles ionizes the gas and the liberated electrons are collected at the anode (Isaksson, 2011a). The registration of alpha particles (or electrons) are measured by an ion current by use of an electrometer. The ionization chamber could be used with flow through or closed operational modes.

Some passive techniques are;

1. **Activated charcoal** is used as an absorber. The charcoal is weighted before measurement and sealed, then exposed to the air that is to be investigated. As soon as measurement is finished, the charcoal is measured again and the radon content is determined by gamma spectrometry.

2. **Nuclear track detector** works on the principal of a selected film material, which due to the release of alpha particles is damaged, where each alpha particles leave a small track. The tracks are enhanced by use of NaOH or KOH to increase the size of the tracks to simplify calculation. The tracks are counted by us of optical micropy or by automated scanning. Knowing the decay rate of $^{220}\text{Rn}$ in relation to the density of the tracks gives an appropriate number of the $^{222}\text{Rn}$ release in the air.

CBI uses a pulsating ionization chamber, called ATMOS 33. The working principle of the instrument and the transformation process of alpha decay converted to radon gas content in air is further explained in Section 5.4.

### 3.2.3 Protection quantities – radon and effective dose

The protection quantity of radon is thus given in Bq/m$^3$, but could also be recalculated to effective dose (UNSCEAR, 2000). In the UNSCEAR report of 2008 (UNSCEAR, 2008) a summary of the total outcome of the effective dose is summarized as

$$E_{\text{eff}} = E_{\text{int}} + E_{\text{ext}}$$  \hspace{1cm} (3.7)

where $E_{\text{eff}}$ = total effective dose from $E_{\text{int}}$ = Internal effective dose produced by inhalation (radon gas) and $E_{\text{ext}}$ = Effective dose external produced by gamma rays.

Estimates of absorbed dose to the critical cells of the respiratory tract per unit $^{222}\text{Rn}$ exposure related to a general population can be derived from analysis of information of aerosol distribution, breathing rate, fractional deposition in the airways, clearance rate and location of the target cells in the airways (UNSCEAR, 2000, Scofield, 1988). However, these different variables are all subject to a certain degree of uncertainty. A simplified relation taking into consideration the above can relate the radon gas content in Bq/m$^3$ to effective dose (UNSCEAR, 2000).

For indoor exposure the relation can be described as;

$$\frac{40\text{Bq}}{\text{m}^3} \times 0.4 \times 7000 \text{ h} \times 9 \text{ nSv} \left(\frac{\text{Bq}}{\text{m}^3}\right)^{-1} = 1.0 \text{ mSv}$$  \hspace{1cm} (3.8)

Where 40 Bq/m$^3$ represents a mean indoors value of the population worldwide, 0.4 represent an equilibrium factor of $^{222}\text{Rn}$ and its related decay products, 7000 hours = 80 % spent indoors annually and 9 nSv = mean average value of dose conversion between inhalation of radon and the above variables.

The range of dose conversion factors for radon, derived from epidemiological studies and physical dosimetry varies from 6 to 15 nSv (UNSCEAR, 1998, 2000). In the report of UNSCEAR (2008) a slightly higher mean value of 10 nSv has been recommended. It should also be addressed that the equilibrium factor may vary largely (0.1-0.9), but as an overall mean it has been justified by the committee (Unscear, 2000).
Nuccitelli et al. (2015) calculated according to formula (equation 3.8) using the mean value of 10 nSv as a mean average instead of 9 mSv. For Swedish regulations a corresponding effective dose$_{\text{ext}}$ of ~6 mSv per year at 200 Bq/m$^3$ of $^{222}$Rn in air would be the result. This is a factor ~6 in effective dose in relation to the reference value of allowed gamma generated effective dose$_{\text{ext}}$ for building materials as presented by CE (2014).

The other factors influencing the total effective dose (E$_{\text{eff}}$), such as ingestion (meaning food/water containing natural radionuclides) are numerically negligible compared to the quantity in effective dose produced by the inhalation of $^{222}$Rn and its progenies (ICRP, 2007, UNSCEAR 2008).

### 3.3 Risk aspects and regulatory framework

#### 3.3.1 Risk aspects of radon gas and gamma radiation

The production of radon gas is an ongoing process in the uranium decay chain (see Figure 2.2), where the $^{238}$Ra atom releases an alpha-particle and hence reduces to the noble gas $^{222}$Rn. The radon gas has a half-life of approximately 3.8 days and its continuing decay results in the release of an alpha-particle and the production of metal-ions, namely $^{218}$Po. Yet further the $^{218}$Po radon-daughter releases another alpha-particle and reduces to $^{214}$Pb and $^{214}$Bi. $^{218}$Po, $^{214}$Pb, $^{214}$Bi and $^{214}$Po are the so called short-lived radon-daughter isotopes (Scofield, 1988). In the process of the decay of $^{218}$Po to $^{214}$Pb and $^{214}$Bi beta- and gamma radiation are also emitted (see Section 2.1, Figure 2.2). Due to the release of gamma radiation between $^{218}$Po and $^{214}$Bi, the $^{214}$Bi – atom is used as a progeny to calculate the activity of $^{226}$Ra assuming a state of equilibrium between $^{226}$Ra and $^{214}$Bi.

The decay of $^{222}$Rn gives rise to a manifold of health aspects. The radon daughters ions namely stick to particles in the air (aerosols) and are inhaled. As such particles in the air (aerosols) are troublesome and different places within a house give rise to different risks (Scofield, 1988, Åkerblom & Clavensjö 2007). For example the kitchen area, when making food, gives rise to numerous particles in the air and as such the risks increase of inhaling larger doses of the daughter isotopes within certain areas of the house (Scofield, 1988). Cigarette smoke and humidity are variables that largely affect the number of aerosols in the air and consequently the risk of bonding with radon and its progenies (Scofield, 1988).

In view of the risks for external exposure alpha-emissions exerted to the human body from the outside could easily be stopped by the skin (Beta particles and gamma rays are though, more deeply penetrating into the skin). But, when $^{222}$Rn and its progenies are inhaled or digested by the food the human cells are more sensitive to the alpha-emission once inside the human body. The ICRP (2007) describes this in terms of an index (W$_R$) correlating to the total equivalent dose or effective dose where alpha-emissions are 20 times as powerful in ionizing its surroundings compared to beta- and gamma rays. In part, due to its charge (W$_R$) but, more importantly its comparatively slow speed giving it time to ionize the atoms more intensely (Isaksson, 2011a, Cember & Johnson (2009)).
3.3.2 Regulatory framework of natural radiation in building materials

Part of the aim of the Thesis is to demonstrate how Swedish concrete, due to different aggregates, is able to fulfill the requirements by the European Commission, within its directive, Basic Safety Standards (BSS), and its article 75, that stipulates the building materials are not to exceed the reference level of 1 mSv per year for building materials (EC, 2014). This excess gamma radiation from building materials (subtracting the outdoors background) are assessed by comparing the reference level of 1 mSv per year in effective dose to an activity index, described as I-index presented by the EC in 1999 (RP 112). Meaning, the I-index (ionizing radiation from $^{40}$K, $^{232}$Th, $^{226}$Ra), shall be less than or equal to 1. The I-index is within this context roughly comparable to 1 mSv per year or less for building materials (Markkanen, 1995, EC, 1996, EC 1999).

The second part concerns the Swedish regulations of radon gas (Swedish National Board of Housing, Building and Planning, 2011), which are stricter than the stipulated EC regulations.

From a regulatory point of view and from a European standpoint the consequence of acquiring a result of a construction product yielding a value > 1 in I-index is not equivalent to a non-usable construction product (EC, 2014). According to the Construction Product Regulation (EC, 2011) and the EU’s BSS and the forthcoming Technical reported presented by the EU working group TC351/WG 3/TG 32, a further assessment of the construction product/building material, however, becomes a necessity. As a consequence, the construction product may still be used and declared in accordance with the CE-marking, however, first after completion of an comprehensive “dose assessment”. The current reference/threshold values from a European perspective as well as on national basis is shown in Table 3.2.

Table 3.2 The reference and threshold values in relation to ionizing radiation of gamma radiation and radon gas.

<table>
<thead>
<tr>
<th>Condition</th>
<th>Level</th>
<th>Unit</th>
<th>Regulation</th>
</tr>
</thead>
<tbody>
<tr>
<td>Levels of ionizing radiation, gamma energies not to exceed 1 mSv per year from building/construction materials expressed as I-index</td>
<td>≤ 1 in I-index*</td>
<td>Dimensionless</td>
<td>Eu’s Basic Safety Standard (BSS)</td>
</tr>
<tr>
<td>Levels of ionizing radiation (gamma energies) within a room</td>
<td>0,3**</td>
<td>μSv/h</td>
<td>Regulations from the Swedish National Board of Housing, Building and Planning</td>
</tr>
<tr>
<td>Levels of radon gas in new buildings</td>
<td>200</td>
<td>Bq/m³</td>
<td>Eu’s Basic Safety Standard (BSS)</td>
</tr>
<tr>
<td>Levels of radon gas proposed from the European Union (BSS)</td>
<td>300</td>
<td>Bq/m³</td>
<td>Eu’s Basic Safety Standard (BSS)</td>
</tr>
</tbody>
</table>

NOTE*: The I-index is a screening tool and the value (I-index) for a building material could be exceeded, but, further dose assessment of the final building material should then be performed as not to exceed the reference level of 1 mSv per year. NOTE**: Most likely, the value of 0.3 μSv/h as a threshold level of gamma radiation within a room may be replaced in the near future as a consequence of the reference level for gamma radiation of 1 mSv per year from building materials.

3 A more comprehensive explanation is given in chapter 4. Methodology – gamma radiation.
4 Methodology – gamma radiation

4.1 General

The activity index, denoted I, is dimensionless. It constitutes a screening tool value in accordance with the EC regulations (EC, 1999) declared in report – RP 112. The EC’s Basic Safety Standard (EC, 2014) further emphasizes that the I-index for building materials should not exceed 1. I-index is calculated as:

\[ I = \frac{C_{40K}}{3000} + \frac{C_{226Ra}}{300} + \frac{C_{232Th}}{200} \] (4.1)

where "C" = concentration in Bq/kg for each radionuclide.

4.2 Test methods - General

Several test methods are applicable for evaluation of the specific activity (activity concentration \(^{40}\)K, \(^{226}\)Ra and \(^{232}\)Th in Bq/kg) of gamma rays. Instruments used in the studies have been a portable hand-held spectrometer in combination with laboratory gamma ray spectrometry, which is a well documented technique (Mauri & Gäfvert, 2013). Laboratory gamma ray spectrometry is also the analytical practice that is recommended by the European Union and its technical committee 351/WG 3 – dangerous substances. The committee has assigned working group 3 (WG 3) and task group 32 (TG32) as to bring forward a proposal/draft of a technical report, as to explain and validate earlier gamma radiation models (Markkanen, 1995, EC, 1999) in relation to building materials.

The work group, WG3, and its task group TG31 of the technical committee have, as of today put forward a draft on measurements of \(^{40}\)K, \(^{226}\)Ra and \(^{232}\)Th using laboratory gamma ray spectrometry and suitable radionuclides for activity measurement. The draft is currently under review. The techniques within this Thesis follow the recommendation set up by the working group and the forthcoming technical report.

The third assessment tool of how to calculate the amount of radioactive elements of natural radionuclides is employed by using geochemistry by ICP-MS/EOS. This is possible by having knowledge of each radioactive elements weight percentage of different oxides.

4.2.1 Test method A – Portable gamma ray spectrometer

The Swedish Cement and Concrete Research Institute (CBI) uses a portable hand-held spectrometer delivered by Radiation Solutions Incorporated.

Measuring by use of scintillation crystals, such as NaOI or BGO-crystals is based on the procedure that the amount of light (number of photons) produced in the scintillation crystal is proportional to the amount of gamma ray energy initially absorbed in the crystal (Knoll, 2010). Hence, also the number of photoelectrons from the cathode, the final anode charge, the amplitude of the preamplifier and amplifier voltage pulses are proportional to the energy absorbed by the crystal (Knoll, 2010). Thus, the instrumentation technique relies on scintillation of a crystal (bismuth germanate oxide), which when exposed to different energies (as photons), will scintillate in response to the incoming energy. The relation of energy could be used to describe the different energies of radionuclide decay, following,

\[ E = hf \] (4.2)

Where, \( E \) = energy in electronvolt often described as keV (kiloelectronvolt), \( h \) = Planck’s constant \((6.63 \times 10^{-34} \text{ Js})\) and \( f \) is equal to frequency of the electromagnetic radiation.
Each gamma ray photon, when interacting with a sensitive crystal, such as NaI or BGO (Bi₄Ge₃O₁₂) crystal (Figure 4.1a, b) generates an optical light pulse (Cember & Johnson, 2009), which will be converted to a specific energy (E) for that specific photon.

Gamma ray photons passing through the crystal interact with the BGO-crystal through photoelectron absorption, compton scattering and pair-production. The primary photoelectrons, electrons and positrons (particles) generated by each process dissipate their kinetic energy by exciting and ionizing the atoms of the BGO-crystal (Cember & Johnson, 2009; Knoll, 2010). The atoms within the BGO-crystal return to their ground state by emission of light ejected to the photocathode (Figure 4.2). The role of the photocathode is to convert the light pulse into an electron (photoelectron), which in turn is amplified by a photomultiplier tube (PMT). The prime objective of the photomultiplier tube is to enhance the electric pulse into a strong signal by multiplying the photoelectrons leaving the cathode. Hence, several highly charge dynodes are used which increase the number of electrons by the principal of multiplication (e.g. one electron hits the first dynode, which ejects two electrons. Two electrons are then ejected to the next dynode, which when ejects four electrons and so forth). As a result, the final dynode ejects numerous photoelectrons, which combined generate a strong current pulse collected at the anode. The anode is attached to a pre-amplifier for further enhancement of the signal (Figure 4.2). The processing of the signal at the end turns out as a readable energy peak (photopake), with its specific energy presented on a chart. Most often the pulse count rate is described as a function of a given energy (Figure 4.3).
Figur 4.2. Description of the scintillation counter and the role of the photocathode, the photomultiplier tube and the anode accumulating the final current pulse. Figure is modified after Marco Gui Alves Pinter’s master Thesis in astrophysics and instrumentation, Colombia, https://estudogeral.sib.uc.pt

The whole principle rests on that the magnitude of the generated pulse (current pulse) is proportional to the energy of the primary ionizing particles charge (Knoll, 2010). Hence, each specific gamma ray photon has its given energy, depending on the radionuclide from which it is emitted. The pulse height spectrum (see Figure 4.3) is a representation of the number of events at a given energy deposited in the detector.

Figur 4.3. Identification of different photopeaks as a function of the photon energy in keV. Inserted in black text are known radionuclides and their typical gamma ray photon energies. To the right is the 1461 keV peak typical of $^{40}$K. Modified after Knoll (2010).

In reality the calculation of the number of pulses (events in the detector) can be distorted by the high number of photons hitting the BGO crystal simultaneously. Hence, there is a limitation called “dead-time”, which works according to the principle of “open or closed” with respect to processing of the signal. Meaning, the time needed for the hand-held spectrometer RS 230 to process a photon interaction is 50 milliseconds, or twenty samplings per second. In Figure 4.4 the three energy windows used to assess the activity of each radionuclides of $^{40}$K, $^{226}$Ra and $^{232}$Th is presented with respect to number of counts within each window as a function of energy. In the software, the defined energy intervals, depicted in green, are used to calculate the area under each peak representing detector events for gamma radiation from each radionuclide ($^{40}$K, $^{214}$Bi and $^{208}$Tl).
Figure 4.4. Location of photopeaks (full energy peaks) as a function of the energy in keV. A logarithmic spectrum from the RS 230 software presenting the energy intervals, in green colour, that are used for calculation of the total energy of \(^{40}\text{K},^{226}\text{Ra}\) and \(^{232}\text{Th}\) respectively. The spread of the full energy peaks are due to statistical fluctuations in the detector and electronics.

Consequently, the incoming photon energy, striking the crystal, initiating scintillation, are at the end described as diverse energy-photopeaks in a spectrum, which is assigned to different elements and their radionuclides decay. Meaning, the radionuclides’ elemental concentrations are directly related to the magnitude (amplitude) of the full energy peaks (counts).

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). When assessing the data, the conversion of elemental concentrations (percent and parts per million) to specific activity (Bq/kg) for each element follows the recommendations of IAEA (1989). Table 4.1 shows the conversion coefficients used.

Table 4.1. The conversion of elemental concentrations to specific activity (IAEA, 1989).

<table>
<thead>
<tr>
<th>Element</th>
<th>Activity Concentration (Bq/kg)</th>
<th>Isotope</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 % Potassium (K)</td>
<td>313</td>
<td>(^{40}\text{K})</td>
</tr>
<tr>
<td>1 ppm Uranium (U, Ra)</td>
<td>12.35</td>
<td>(^{238}\text{U}, \text{or}^{226}\text{Ra})</td>
</tr>
<tr>
<td>1 ppm Thorium (Th)</td>
<td>4.06</td>
<td>(^{232}\text{Th})</td>
</tr>
</tbody>
</table>

RS-230 measures, as stated, the gamma radiation energies, in keV from the decays of \(^{40}\text{K}\) (1461 keV), \(^{214}\text{Bi}\) (1760 keV) and \(^{208}\text{TI}\) (2615 keV) in three different energy windows, 1370-1570 keV, 1660-1860 keV and 2410-2810 keV respectively, also shown in Figure 4.4 and tabulated in Table 4.2.
Table 4.2 Natural isotope of interest and energy intervals that are used to calculate the activity of the primordial nuclides. The energy peaks are defined as well as the energy range used to calculate the different dose rates in either the unit (µSvh/h) or (nGy/h).

<table>
<thead>
<tr>
<th>Primordial nuclides</th>
<th>Isotopes used to measure</th>
<th>Peak energy of measured isotope (keV)</th>
<th>Energy interval (keV) used in calculation of dose rate/h</th>
</tr>
</thead>
<tbody>
<tr>
<td>40K</td>
<td>40K</td>
<td>1461</td>
<td>1370 – 1570</td>
</tr>
<tr>
<td>226Ra</td>
<td>214Bi</td>
<td>1760</td>
<td>1660 – 1860</td>
</tr>
<tr>
<td>232Th</td>
<td>208Tl</td>
<td>2615</td>
<td>2410 – 2810</td>
</tr>
</tbody>
</table>

By use of regions of interest (ROI) as defined by the energy windows in the Table 4.2, the total count rate are performed according to relationships of Exposure (X), absorbed dose in air (D) and ambient dose, H*(10). Table 4.3 gives the relations to calculate the total count rate in Röntgen (R), absorbed dose rate in air (D) and the approximate relation to ambient dose rate, H*(10).

Table 4.3 Conversion factors, established 1 m above ground between the total exposition in röntgen (R), absorbed dose rate in air (D) or the ambient dose rate (uSv/h) using conversion factors of IAEA (2003).

<table>
<thead>
<tr>
<th>Primordial nuclides</th>
<th>Exposure rate (uR/h)</th>
<th>Absorbed dose rate in air (nGy/h)</th>
<th>Ambient Dose rate H*(10) – uSv/h¹</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 % Potassium (K)</td>
<td>1.505</td>
<td>13.078</td>
<td>~0.0154</td>
</tr>
<tr>
<td>1 ppm Uranium (U, Ra)</td>
<td>0.653</td>
<td>5.675</td>
<td>~0.0668</td>
</tr>
<tr>
<td>1 ppm Thorium (Th)</td>
<td>0.287</td>
<td>2.494</td>
<td>~0.0293</td>
</tr>
</tbody>
</table>

¹) Depends on soft tissue and energy of photons (in keV).

Calculation of each radionuclides’ activity contribution (Bq/kg) to the total count rate (µR/h, n/Gy or µSvh/h) is achieved by multiplication of the conversion factor of each nuclide respectively (Table 4.3). As an example,

2.4 % of K is measured by the field spectrometer. To calculate the corresponding dose in absorbed dose rate in air (nGy/h), 2.4 % × 13.078 (Table 4.3) = 31.39 nGy/h.

The respective counts of each nuclide, 40K, 226Ra (214Bi) and 232Th (208Tl), are multiplied in the same manner as the example above. Finally, each radionuclides’ count rate is added together and represents the total counts/h. Hence, and worth noting, is that in relation to some other spectrometric devices, the whole energy field from 30 to 3000 keV is not surveyed by the RS 230-instrument.

The count rate could also be controlled by use of the activity concentration in Bq/kg converted to ambient dose rate (µSvh/h) according to tabulated values in the report of The Radiation Protection Authorities in Denmark, Finland, Iceland, Norway and Sweden (2000). In Table 4.4 is given the ambient dose equivalent rate conversion from nSv per Bq/kg for each radioisotope. A crosschecking with the instruments readout in µSvh/h was also performed in relation to the tabulated values. The instrument yields a good correlation, however, slightly higher numerical coefficient values have experimentally been determined for the ambient dose rate, when measured on a plane surface.
Table 4.4 Ambient dose equivalent rates and their conversion coefficients according to tabulated data as well as experimentally determined with the RS 230 at a dose rate of ~0.1 µSv/h ambient equivalent dose rate.

<table>
<thead>
<tr>
<th>Radioisotope</th>
<th>Ambient dose equivalent rate, nSv/h per Bq/kg (1m above surface)</th>
<th>Ambient equivalent dose rate - numerical values, experimentally determined for the RS 230, nSv/h per Bq/kg (at the surface)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{226}\text{Ra}$ ($^{238}\text{U}$)</td>
<td>0.531</td>
<td>0.540</td>
</tr>
<tr>
<td>$^{232}\text{Th}$</td>
<td>0.718</td>
<td>0.0741</td>
</tr>
<tr>
<td>$^{40}\text{K}$</td>
<td>0.049</td>
<td>0.052</td>
</tr>
</tbody>
</table>

The conversions from the measured radionuclides to total concentrations of potassium, uranium and thorium are based on natural occurrences of the isotopes in the earth (e.g. $^{40}\text{K}$ constitutes 0.0117 % of natural potassium). Moreover, in order to calculate concentrations of $^{226}\text{Ra}$/$^{238}\text{U}$ and $^{232}\text{Th}$ from the daughter isotopes $^{214}\text{Bi}$ and $^{208}\text{Tl}$, radioactive equilibrium in the decay chains is assumed (i.e. no element has been added or removed).

In case of radioactive (secular) equilibrium, the activity concentration of all members of the decay chain is equal (Turner, 2012). Hence, $^{214}\text{Bi}$ can be taken as a proxy for both $^{238}\text{U}$ and $^{226}\text{Ra}$ and $^{208}\text{Tl}$ can be taken as a proxy for $^{232}\text{Th}$ (see Figure 4.5). It can be shown that in about 7 half-lives of the daughter, the condition of secular equilibrium reaches a steady state where the parent atoms activity and its decay rate is equal to its daughters activity and decay rate by the expression $A_2 = A_1$ (Figure 4.5). It could also be addressed in relation to the parents and daughter decay rate and number of atoms (N) as:

$$\lambda_1 N_1 = \lambda_2 N_2$$  \hspace{1cm} (4.3)

The influence of measuring on daughter isotopes, such as $^{214}\text{Bi}$, is well demonstrated by Mauring & Gäfvert (2013) and shows the importance of sealed samples, to make use of this relation, especially for the relation of $^{226}\text{Ra}$.

![Figure 4.5 State of of equilibrium between different isotopes in a decay chain expressed as “activity (Bq) as a function of time”. When the build up in activity of the daughter isotope (A2) has reached ~7 half -lives the activity of the parent (A1) = daughter (A2) and secular equilibrium is said to exist. Figure modified from Cember & Johnson (2009).](image-url)
A yearly calibration of the hand held gamma spectrometer has been performed at the national calibration site for field gamma equipment, situated at Borlänge airport (see Figure 3.2).

4.2.2 Test Method B – Laboratory gamma spectrometry

Advanced gamma spectrometric analyses, which is used by The Radiation and Nuclear Safety Authority of Finland (STUK) practices an accredited testing method - gamma-spectrometry, in-house guide VALO 4.5 (Klemola et al., 2010) using semiconductors with High Purity Germanium Detectors (HPGE). The advantage of using semiconductors lies in their ability to carry large numbers of charge carriers, which gives a good energy resolution (Knoll, 2010). As a consequences of the large number of carriers per pulse the statistical error of the detector becomes very small. Secondly, their sensitivity to ionization energies is very small compared to other techniques (for Germanium ~3 eV).

The technique relates to the use of the configuration of the bonding of the atoms (lattice) and their internal distance within crystals, e.g. germanate or silica. Electrically, solids can be classified as insulators, semi-conductors or conductors (Turner, 2012). The lattice in a crystal could be described in terms of valence band and conduction bands, where a distance between the two layers are defined as the band gap (Knoll, 2010). This is demonstrated in Figure 4.7a. In order to make a current flow, one has to overcome the energy needed as to make electrons move across the band gap (Isaksson, 2011a).

As a mean to achieve this objective, with as little energy input as possible, “dopants” are used. Dopants are other atoms, closely associated in their atomic structure with the atoms used in the crystal. They are considered as “impurities” in the crystal (Knoll, 2010). Impurities are added as part of the melt, when forming new crystals (Turner, 2012). By adding arsenic (As) as a dopant in a germanium (Ge) melt one will create an excess of electrons in the crystal, and hence an n-type semi-conductor as shown in Figure 4.7b are produced. The p-type semi-conductor is doped in the same manner but in this case with an element that has atoms with fewer electrons in their outer shells then required for a covalent bond in the crystal to form (Turner, 2012). Consequently, due to its shortage of electrons between some atoms, it will be positively charged.

For radiation purposes, in order to register the photon interaction by a crystal, a very small bandgap is desired. The photon interaction from a sample, will in the active detector (germanium crystal) release electrons, which can be collected if a small external current is applied (Isaksson, 2011a). This process, to register an electric charge, is as described, aided by dopants. The energy released by the crystal as a pulse is proportional to the electric charge of the ionizing photon (Turner, 2012).

But, striving for a very sensitive detector, aided by dopants, also means, that the crystal becomes very sensitive to changes in temperature (thermal heat), which will make electrons move across the band gap (Knoll, 2010, Isaksson, 2011a). The thermal influence on the semiconductors is often defined as noise (or current leakage) and its suppression is essential when using semiconductors (Knoll, 2010). In order to reduce the noise a constant temperature and constant cooling constitute a necessity as to ensure good reliability. The coolant is almost always liquid nitrogen.
Thus, a configuration using a detector with germanium automatically implies cooling with liquid nitrogen. In Figure 4.8a different dewars used for cooling of the detector are shown.

Figure 4.8a. Different dewars from Canberra mounted with detectors. Published with courtesy of Canberra Inc.

Figure 4.8b. A lead and copper shielded chamber for measurements. Model 767 – Canberra. At the bottom of the container a hole can be seen where the detector is inserted (red arrow). Published with courtesy of Canberra Inc.

Figure 4.8c. Illustration of the detector (grey) when measuring a sample (contained in the Marinelli beaker) and shielded from external influences as shown in Fig. 4.8b.

The same principle in enhancing the signal output applies as for method A, where a preamplifier often is mounted close to the detector as to enhance the signal output. The height of the energy peak (count rate) is thereafter defined in relation to the specific magnitude of the charge (keV) induced by the interacting photon (Knoll, 2010).

At STUK, the detectors are placed in cylindrical background shields made of 120–140 mm thick lead. The shields are lined inside with cadmium and copper (Figure 4.8b). The measured energy range is 30–2700 keV. The relative efficiency of the detectors varies from 37 to 90 % and with energy resolution between 1.6 keV and 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, sealed with a vacuum-tight aluminum folie in order to achieve equilibrium. For the measurements performed for CBI two
spectrometers have almost always been used: F5 (Ortec GEM-35190) with an energy resolution of 1.70 keV at 1.33 MeV and a relative efficiency of 37.6 % and G3 (Ortec GEM-30P4-S) with an energy resolution of 1.76 keV at 1.33 MeV and a relative efficiency of 40 %. Their resolution and relative efficiencies are rather similar and cross-references between the detectors have been made with very good reliability and trustworthy results (personal communication Seppo Klemola, 2012).

AT STUK, the gamma lines for assessing the three radioactive nuclides are measured 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. Also, in this case, secular equilibrium has to be assumed for $^{228}$Ac and in part for the correct assessment of $^{226}$Ra peak. For calculation of the photopeaks and their intensities, specific attenuation has to be made for the $^{226}$Ra peak (186.2 keV), where comparative data also include crosschecking with the $^{235}$U-peak (185.7, 143.8, 163.4 keV photopeaks). Also, cross-checking with $^{214}$Bi and $^{214}$Pb (assuming secular equilibrium) aids in deciding the correct photo peak height (energy) for $^{226}$Ra. For $^{232}$Th, the $^{228}$Actinium photopeak is more easily measured with less intense photopeaks nearby creating interference, but assumes conditions of secular equilibrium. The $^{40}$K photopeak is measured directly and is normally strong and less sensible to influence from other nearby photopeaks.

The correction for sample height and density, as well as the effect of true coincidence summing$^4$, is also taken into account in the calculation of the results. The uncertainties include both statistical uncertainty and uncertainty due to the efficiency of calibration (Klemola et al., 2010).

4.2.3 Test Method C – Geochemistry and ICP-MS/EOS

Inductively Coupled Plasma/Optical Emission Spectrometry (ICP-OES) is used to measure major and rare earth elements. The laboratory, CRPG/CNRS makes use of an ICP-OES called Thermo Icap 6500 with a radial torch.

The technique relies on that excited atoms and ions in a discharge plasma, creating a unique emission spectrum specific to each element. ICP-OES measures the light emitted at element-specific characteristic wavelengths from thermally excited ions. This light emitted, through excitation, is separated and measured in a spectrometer, yielding an intensity measurement that can be converted to an elemental concentration by comparison with calibration standards. Figure 4.9 shows a simplified scheme of the principle.

$^4$ At high count rates, the limited time resolution of the detector system may result in two emitted photons being registered at the sum of their individual energies. This phenomenon is denoted true coincidence summing and results in an additional sum peak and a decrease in the peaks corresponding to the individual photon energies.
Figure 4.9. The working principle of an Inductively Coupled Plasma/Optical Emission spectrometer, where the sample in the plasma generates a unique emission spectrum for each element as illustrated to the right by different elements at different wavelengths. The figure is modified from www.chemwiki.ucdavis.edu.

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).

The ICP-MS measures the masses of the element ions generated by the high temperature argon plasma. The ions created in the plasma are separated by their mass to charge ratios, enabling the identification and quantitation of materials. In Figure 4.10 the principle of a mass spectrometer is shown. To the left in the Figure the sample is vaporized (nebulized) before entering the torch and ionized by the argon plasma. The ions are collected and transported to the quadrupole, where they are accelerated to the detector. The detector registers an electron, which is then amplified by dynodes, in the same manner as for method A as to get a strong electric pulse, which could be identified as a peak energy for that specific element.

Figure 4.10. The principle of a mass spectrometer. To the left the sample is vaporized (nebulized) before entering the torch and ionized by the argon plasma. The ions are collected and transported to the quadrupole, where they are accelerated to the detector. Figure modified from www.eecelabs.seas.wustl.edu
The results for potassium (K) have been reported as potassium oxide (K₂O). A correction for 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.

4.3 Assessed materials

4.3.1 General overview

The area of interest has been concentrated to the two major cities of Sweden; Gothenburg and Stockholm, including their suburbs and the area between the cities. Due to the large demand of crushed bedrock (aggregates) within these regions with regard to numerous infrastructural and housing projects the natural outcome resulted in a focus on this area. The bedrock is between these two major cities partially enriched (I-index > 1) in natural radionuclides.

A compilation of the natural radioactivity of bedrocks across Sweden has been presented by the Swedish geological survey (Jelinek and Eliasson, 2015). Jelinek & Eliasson (2015) made use of aerial data between the years 1969 and 2014. Part of this data is presented in Figures 4.11a and 4.11b.

In relation to these aerial data a simultaneous compilation of spectrometric surface data was assessed indicating that almost ~25% of the spectrometric readings, when recalculated to I-index, exceeded the screening tool value of 1. In Figure 4.11b some of these elevated areas are presented in conjunction with the project area assessed in this Thesis.

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Fig 4.11a. Aerial data of the gamma radiation expressed in the dimensionless unit I-index (AI-index). Copyright SGU.

Fig 4.11b. Aerial data of the gamma radiation presented as I-index (AI-index). The brown areas are highlighted as slightly elevated, where radiation is close or above an I-index of 1. The encircled area is the project area where the quarries and the examined bedrock/aggregates originate. Copyright SGU.
4.3.2 Aggregates, concrete specimens and samples

The study included crushed rock aggregates (Table 4.5), which entail almost the full range of variations in natural radiation from gneisses and granitoids known within Sweden. 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 (Figure 4.11b).

Twelve different aggregates have been examined. In one case, the same aggregate has been used to make concrete where different w/c-ratios have been applied (sample 7A, 7B, Table 4.5). The initial conditions of the concrete recipe applied a moderate to low w/c ratio of 0.45.

Table 4.5. Different crushed rock materials/aggregates as well as the cast concretes and their thicknesses. References to appended papers are also given. Sample 7 was cast with two different w/c ratios. In the current papers, reference is to the cast specimen with w/c ratio of 0.45

<table>
<thead>
<tr>
<th>Sample</th>
<th>Concrete samples used for radon gas evaluation, phase I, chapter 7</th>
<th>Bedrock</th>
<th>Concrete thickness (mm)</th>
<th>Paper No.</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>(5) Granitoid</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>(2) Granitoid</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>(1) Granitoid</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>(7) Granitoid/gneiss</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td></td>
<td>Gneiss</td>
<td>150</td>
<td>I, II and III</td>
</tr>
<tr>
<td>6</td>
<td></td>
<td>Gneiss</td>
<td></td>
<td></td>
</tr>
<tr>
<td>7A</td>
<td></td>
<td>Granitoid</td>
<td></td>
<td></td>
</tr>
<tr>
<td>8</td>
<td></td>
<td>Gneiss</td>
<td></td>
<td></td>
</tr>
<tr>
<td>9</td>
<td></td>
<td>Granitoid</td>
<td></td>
<td></td>
</tr>
<tr>
<td>10</td>
<td></td>
<td>Gabbro</td>
<td></td>
<td></td>
</tr>
<tr>
<td>11'</td>
<td></td>
<td>Granitoid</td>
<td>150</td>
<td>200</td>
</tr>
<tr>
<td>11''</td>
<td></td>
<td>Granitoid/gneiss</td>
<td>150</td>
<td>200</td>
</tr>
<tr>
<td>13</td>
<td></td>
<td>Granitoid</td>
<td></td>
<td>III</td>
</tr>
<tr>
<td>14</td>
<td></td>
<td>Granitoid</td>
<td></td>
<td>III</td>
</tr>
<tr>
<td>15</td>
<td></td>
<td>Granitoid</td>
<td></td>
<td></td>
</tr>
<tr>
<td>16</td>
<td></td>
<td>Granitoid</td>
<td></td>
<td></td>
</tr>
<tr>
<td>17</td>
<td></td>
<td>Granitoid</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Note 1) Same aggregate as sample 3. 2) Same aggregate as sample 4.

The project covers all in all eighteen full scale concrete slabs (1500 × 1500 mm in length and width), constituting eleven specimens with a thickness of 150 mm, four specimens with a thickness of 200 mm as well as three specimens, with 100 mm in thickness.

However, as the project progressed, a few concrete samples (300 × 300 × 150 mm) were also cast with a w/c ratio of 0.65 (phase II). This was predominantly done in relation to investigation of radon gas. Finally, a third set, constituting two concrete specimen of 300 × 300 × 150 mm in size, with different w/c ratios (0.65 and 0.45) were cast, using the same aggregates, from one quarry, also assessed in phase II.
One of the large concrete castings (1500 × 1500 × 150 mm) was produced by a ready-mix concrete plant. Two particle size fractions were used; 0-8 and 8-16 mm. The recipe should be in accordance with the actual sieve curve used in the project (personal communication – Per Malmbäck/CBI).

4.3.3 Concrete recipe

The concrete specimens have been produced by mixing two or three aggregate particle size fractions. In one case, one particle size fraction (0/18) was used. A standard 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 as binder. The component of air was set to approximately 1.5 %. A cement-content of 350 kg/m³ was applied. For concrete used for dwellings a minimum strength class of at least C30/37 was set as the primary target (Table F.1, SS-EN 206). Using the current w/c ratio, a strength development of approximately 55 MPa (28 days) could be anticipated for the concrete specimens. For the concrete samples with a w/c ratio of 0.65, primarily investigated for radon gas exhalation, the minimum strength class was set to C25/30. Additives (superplasticizer) were used in a minor portion to have a slump of approximately 100 mm for good workability of the concrete. Table 4.6 shows an example of the general recipe of the concrete specimens using a w/c ratio of 0.45.

Table 4.6. General proportions (in kg/m³ and weight %) of the different constituents in the concrete specimens.

<table>
<thead>
<tr>
<th>Constituents</th>
<th>kg/m³</th>
<th>Weight (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cement, CEM II</td>
<td>350</td>
<td>14.9</td>
</tr>
<tr>
<td>Crushed, aggregate, 0/8 (42 wt%)*</td>
<td>770</td>
<td>32.9</td>
</tr>
<tr>
<td>Crushed, aggregate 8/16 (58 wt%)*</td>
<td>1064</td>
<td>45.4</td>
</tr>
<tr>
<td>Water</td>
<td>160</td>
<td>6.8</td>
</tr>
<tr>
<td>Air</td>
<td>0.01 (~1.5 %)</td>
<td>-</td>
</tr>
<tr>
<td>Superplasticizer (Sikament 56)</td>
<td>~1</td>
<td>-</td>
</tr>
<tr>
<td>Total</td>
<td>~2347</td>
<td></td>
</tr>
<tr>
<td>w/c ratio</td>
<td>0.45</td>
<td></td>
</tr>
</tbody>
</table>

*Note, all the aggregates particle size fractions for each concrete cast are presented in Paper III.

4.4 Process of sampling

4.4.1 Aggregates

In order to have similar conditions and to achieve good workability of the concrete, a reference aggregate sieve curve prescribed in SS-EN 1766 was used as a template before mixing the concrete. Consequently, the particle size fractions (weight %) of the aggregates (e.g. example 0-4, 0-8 and 8-16 mm) between the different recipes have been similar. In one case a slightly more coarse material was used due to the crushing as one fraction (0-18 mm in particle size fraction). The initial sampling of delivered aggregates was made according to the standard SS-EN 932-1 where the fractions were collected from different depths and units of the delivered woven bag (Fig 4.12a).

A total weight of approximately 30 kg for each particle size fraction was selected and blended (weight %) according to the prescribed sieve curve (SS-EN 1766) for each concrete recipe. The blended
aggregates were then crushed in a cone crusher (Figure 4.12b) yielding particle size fractions of 0-6 mm. Finally, in order to reduce the sample weight (size), a rotational splitter (SS-EN 932-2) was used (Figure 4.12c). Between 6 and 8 kg were sent for gamma spectrometric analysis (Figure 4.12d). The aggregate mixtures where denoted \( A_{\text{mix}} \), specifying that several particle size fractions were blended.

![Image](image1.png)

Figure 4.12a, b, c, d from upper left two lower right, (a) delivered material in woven bags, (b) cone crusher used to crush sample to fraction 0-6 mm, (c) splitter used to reduce sample size, (d) material after crushing with a polished Swedish coin (krona) used as reference.

In a few cases a different procedure was used. For some aggregates each particle size fraction was sent for analysis. Thus, 30 kg of each particle size fraction were dug in a random fashion from the materials delivered in the woven bags. The procedures to crush, homogenize and reduce the sample weight were identical as for the \( A_{\text{mix}} \) samples.

As a consequence of the latter sampling technique, the final activity concentrations of \( ^{40}\text{K} \), \( ^{226}\text{Ra} \) and \( ^{232}\text{Th} \) of the aggregates (e.g. 0-8 and 8-16 mm) were calculated knowing each particle size fractions activity concentration (Bq/kg) and adjusted, due to its percentage (weight \%) according to the used sieve curve (SS-EN 1766) for that specific concrete recipe (see Section 6.7 for calculation). These aggregate samples are denoted \( A_w \) (Aggregate weighted).
4.4.2 Concrete

Initial casting (Figure 4.13a, b) was done in wooden frames.

Figure 4.13a. CBI employees prepare for casting of a concrete specimen. A lift fork is used to assist before the concrete is poured into the mold. The inner diameter of the wooden frame is 1500 × 1500 × 300 mm in height.

Figure 4.13b. CBI employees pour the first load of concrete into the mold.

Figure 4.14a. The concrete specimen, two weeks after molding and with its covering cloth removed. The casting is still wet in spots and rests within its wooden mold/frame. Blue lines were drawn from the centre of the concrete specimen as to obtain four identical samples of 300 × 300 mm in width and length.

Figure 4.14b. The concrete specimen during measurement by the RS 230. The centre is analyzed.

After initial molding the cast specimens were cured at the surface, to avoid cracks, by using a large cloth that was kept wet. On top, a plastic foil was put to maintain a high degree of moisture.

After the concrete specimens had been allowed to cure for two weeks to gain strength the cloth was removed. The first step of the investigation was to locate the centre of the specimen. Out from the centre, four identical sample squares were drawn (Figure 4.14a,b). Thereafter, from the centre and to each of the drawn sample squares outer “edges” marks were made with a pen. The marks were made 200 mm apart yielding three marks within each sample square (including the centre point).
The gamma spectrometric analysis, as described in Figure 4.14b was conducted at 4 × 3 locations resulting in a combined mean result of twelve readings of four minutes each. All in all, four measurements were made at the centre combined with two measurements within each minor sample (eight readings).

After measurements of gamma radiation the large specimen was sawn along the blue lines (Figure 4.14a). Thus four identical samples of the concrete specimen were stored for further use and complementary measurements by analytical gamma spectrometry, geochemical analysis and radon gas evaluation (Figures 4.15a, b, 4.16a, b).

Fig 4.15a. Cast concrete specimen with the drawn lines and dots marking the four identical squares in size. Fig 4.15b. Four concrete samples sawn from the large concrete specimen.

Figure 4.16a. Quadratic samples of 300 × 300 × 150 mm in size cut from the large concrete specimens after gamma spectrometric measurements. Figure 4.16b. Sawn concrete samples resting beside each other. In the picture the crosses marked by red shows the measurement points used for measurement by the field spectrometer.

For further evaluation of the concrete’s characteristics and its content of the natural radionuclides \(^{40}\text{K}, ^{226}\text{Ra}\) and \(^{232}\text{Th}\), as well as content of radon gas \(^{222}\text{Rn}\) two out of the four sawn samples of the concrete specimens (1500 mm × 1500 mm) were chosen.
The quadratic samples were randomly chosen and cut in half. One half from each samples, once again randomly picked, was selected for crushing. The crushing was done in two steps by laboratory crushers. After crushing of the concrete material the particle size fraction of 0-6 mm dominated. The crushed portion of the samples was subsequently, once again, reduced in size by a rotational splitter according to SS-EN 932-2. A final portion of ~6-7 kg was sent for gamma spectrometric analysis as well as geochemical analysis by use of ICP MS/EOS. The portions were sent to STUK and GRPG/CNRS to compare the results between the laboratories as well as to check the correlation with the result produced by the gamma field spectrometer. Multiple gamma spectrometric analyses of the same portions (blind tests) were also conducted to check the repeatability of the results.

4.5 Rapid determination of concrete samples using field gamma spectrometer

4.5.1 General

The methodology relies on the assessment of the results measured by the field gamma spectrometer on the large concrete specimen in relation to the measurements of the sawn samples originating from the same concrete specimen. The procedure is an estimate and serves a purpose as a rapid analysis and for the further assessment of the concrete’s radon gas exhalation rate. The methodology makes use of a specified recipe. An estimate of the I-index has been the main objective.

4.5.2 Measuring procedure on small concrete slabs.

One of the four sawn concrete samples selected out of the larger concrete specimens was randomly chosen for assessment.

A concrete box (Figure 4.17a, b), approximately 1000×700×700 mm was cast using bedrock with defined low radioactive content of the natural isotopes. The thickness of the sides of the concrete box was approximately 150 mm and with a concrete top/lid of equal thickness. On the inside of the concrete box, the walls and floor is lined with 30 mm thick lead cover for shielding of terrestrial and cosmic radiation (as was described in Section 2.9). At the top double layers of 10 mm thick lead lids were placed before the concrete lid of 150 mm is placed to make a complete enclosure of the lead lining. Using lead as an absorber as explained in Section 2.2.2 reduces the background and cosmic ray influence significantly.

Measurements were performed in the free air (outside) to limit any increased contribution of terrestrial background contributions most often obtained within buildings.

In order to assure as good quality as possible, the measurements made on the sawn concrete samples were made for a minimum of 15 minutes with the portable hand-held spectrometer. The spectrometer was always placed with its BGO-crystal (at the front of the instrument, below the digital display) at the centre of the concrete sample.
Figure 4.17a. Concrete box with a concrete lid swinging in the air by use of a fork lift. The concrete samples used for evaluation of the total activity (Bq/kg) within each concrete mix are shown in the background.

Figure 4.17b. Set up before initiation of the measurement. The lead sealing can be seen inside the concrete walls, as well as the sample with the portable field equipment resting on top of the sample. The portable equipment is, however, not positioned correctly as appropriate when assaying.

4.6 Limitations of measurements and sampling

A part of the investigation is to compare different results and cross-correlate. Measurements of gamma energies can with good reliability be achieved in laboratories; however, the results could be addressed as indefinite, due to how the aggregates have been analyzed. The finer particle fraction that has been analyzed, the more likely it is for a higher activity concentration of the radioactive isotopes, due to increase of particle area/surface within a given volume (Møre, 1985). Within this project all aggregates have been homogenized and crushed to a fraction of 0-6 mm, most likely revealing an excess of ionizing radiation due to the fineness of the aggregates in relation to a particle size fraction as 8-16 mm. The concrete specimens shield gamma radiation in a process called attenuation or self-absorption. Different elements with radioactive nuclides, e.g. \(^{40}\)K, \(^{232}\)Th, \(^{226}\)Ra have different attenuation coefficients as described in Section 2.2.3. The different measurement techniques is an important tool to assess the results correctly and the reader is advised to bear in mind the difficulties and the measurement uncertainties that always accompanies the results. This is addressed in Paper I, where differences in activity of some samples are highlighted.
5 Methodology – radon gas

5.1 General

The purpose of the measurements is to clarify whether the relative humidity (RH) and ultimately, the water content (moisture content) of the concrete have an influence on the radon exhalation rate. At the end, this may influence the final radon content in Bq/m$^3$ of air within a defined space (room).

According to Ishimori et al. (2013) there is a clear relation between increased moisture content and an increase in radon emanation of soils. The variation of the emanation coefficient ($E$) in relation to humidity can for soils also be expressed by the radon production rate per unit volume, $P$ (Bqm$^{-3}$s$^{-1}$)

\[ P = \lambda RE \rho_b \]  

where, $\lambda = \text{decay constant for radon} \ (\text{s}^{-1})$, $E = \text{emanation coefficient (dimensionless)}$, $R = \text{Radium activity concentration in the material (Bq/kg)}$ and $\rho_b = \text{bulk density (kg/m}^3\text{)}$ of the material.

The increase in the production rate at water saturation is due to the presence of water between the pore spaces (Ishimori et al., 2013). The water hinders radon to access the adjacent grains, when decay occurs and the recoil energy of the atoms in the $^{238}\text{U}$-chain is more likely to terminate its recoil in the water. Hence giving rise to an increase of $^{222}\text{Rn}$ release in the water. Consequently more atoms exist in the water and are exhaled into the air as excess water in the poor system evaporates (Ishimori et al., 2013). At a moisture content less than 3 % (weight) or 5-7 % (volume), there is a rapid decrease of the emanation coefficient.

However, increased moisture content is also controlled by the diffusion coefficient (m$^2$s$^{-1}$), which describes the movement of the radon atoms within the material/aggregates and pore spaces. The higher degree of saturation of water, the slower the atom moves and less radon atoms reach the surface of material/aggregates. The diffusion coefficient in soil is largely related to water saturation factor and varies with a factor of ~4 in magnitude from dry to water saturated soils (Ishimori et al, 2013.).

Thus the exhalation of radon gas is largely dependent on the following variables (Ishimori et al., 2013);

1. $R$, the radium activity concentration (Bq/kg)
2. $\rho_b$, the bulk density of the material (kg/m$^3$)
3. $E$, the emanation coefficient (non-dimensional)
4. $D$, the diffusion coefficient of radon (m$^2$/s)
5. $Z$, depth of material

For concrete the conditions of radon exhalation rate are related to the hydration process of the concrete with gel-pores and capillary pores as a function of time (van der Graaf & Meijer, 2003). As long as free excess of water exists, described by Samuelson (1990) the dominating process for concrete seemingly is the emanation coefficient, which largely affects the exhalation rate of radon gas. This is also shown by Yu et al. (1997) who examined the effect of the curing period of some concretes. The exhalation rate of radon gas from the surface of the concretes was initially almost always higher during the first 3 months of examination.

Van der Graaf & Meijer (2003) made several investigations with concrete varying the moisture content while measuring the radon emanation factor and the exhalation rate of concrete. Several concrete specimens, using a w/c ratio of 0.48, were cast and measured in relation to moisture, relative humidity, mass and exhalation rate.
The emanation factor increased linearly for the investigated concretes as the moisture content was increased. For a natural sand the behaviour was quite different, reporting constant values above 5 % in moisture content.

The result in relation to exhalation rate and relative humidity indicates a maximum level of radon exhalation at ~80 RH and thereafter it steadily drops as RH decreases. Between RH of 80 to 90 % there is a sharp decrease of the exhalation rate (Figure 5.1).

![Figure 5.1 Calculated exhalation rates of concretes according to van der Graaf & Meijer (2003).](image)

De Jong & van Dijk (1996), who investigated several types of concrete specimens and combinations with blast furnace and fly ash, reported strong, but in part imprecise results. The emanation coefficient, demonstrated as emanation power, was shown to have a strong relationship to the water content of the concrete samples. For a concrete where the exhalation rate was measured as a function of curing time, the results indicated fairly constant values (but the exhalation rates measured have to be seen as very low). But, on the other hand, some concrete samples showed an increase of the exhalation rate by increasing the cement content, but also simultaneously increasing the free excess of water (w/c ratio 0.53 used), non-related to the type of concrete. This was explained as increment of capillary pores (more spaces within the concrete) giving space for free radon atoms to migrate. Consequently as a result of more capillary pores, the exhalation rate of radon may increase. A similar result was achieved by raising the water content, and consequently, the water cement ratio (w/c-ratio), which according to Reinhardt (1985) in turn yields an increased pore volume and consequently a higher amount of radon release in the pore volume. The results were in line with earlier results reported by Kunsch (1989).

According to de Jong & van Dijk (1996), a lower w/c ratio would in general generate a lower exhalation rate due to less capillary pores and no room for $^{222}\text{Rn}$ to escape into the free air. Secondly, a higher w/c-ratio may initially lead to a higher emanation factor and exhalation rate.

A comprehensive study performed by the SP Swedish Technical Research Institute (Petterson et al, 1982) also reveals that the exhalation rate could vary strongly due to the moisture content of the concrete. The results according to Petterson et al. (1982) implied an approximate increase of the radon exhalation rate of ~10 % in the range of 2.5 – 4 % in moisture content, equivalent to ~50-90 % in relative humidity. Meaning, the trend was similar to the one demonstrated by van der Graaf (2003).

There are also different methods to reduce the radon gas within concrete by substitution of the binder cement. Chauhan & Kumar (2013) have for concrete showed the effect of using silica as a way to reduce the radon exhalation rate of radon. Similar results have been presented by Taylor-Lange et al.
Keller et al. (2001) using different amounts of fly ash in the concrete. Keller (2001) related his results to a better compaction effect by using fly ash in combination with cement and consequently less pore space for free radon transport within the concrete to the surface, reducing the radon gas contribution in air.

5.2 Assessed materials

Initially a set of nine different concrete samples were chosen as parts of larger concrete specimens. The concrete specimens were cast with a w/c-ratio 0.45 (as often within the precast concrete industry).

In addition five more samples were cast with different aggregates and in two out of five cases; the aggregate content was identical to the first eight concrete samples. These five concrete samples had a w/c-ratio of 0.65 (phase II). This later addition of five concrete mixes was propelled in order to investigate a higher w/c-ratio of concrete due to its purpose as building material within a house.

Thirdly, two more concrete cubes (300 × 300 × 150 mm) were cast (phase III), using the same aggregate, but with different w/c ratios.

Table 5.1 shows the different materials and their combinations of aggregates and different w/c ratios.

Table 5.1 Different concrete samples, set/phase and w/c ratios as well as combination of particle size fraction, cement content and water added.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Set/phase</th>
<th>w/c</th>
<th>Aggregate mix (mm)</th>
<th>Cement content</th>
<th>Water</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>I</td>
<td>0.45</td>
<td>0-8 + 8-16</td>
<td>~350</td>
<td>~158</td>
</tr>
<tr>
<td>B</td>
<td>I</td>
<td>0.45</td>
<td>0-18</td>
<td></td>
<td></td>
</tr>
<tr>
<td>C</td>
<td>I</td>
<td>0.45</td>
<td>0-4 + 6-16</td>
<td></td>
<td></td>
</tr>
<tr>
<td>D</td>
<td>I</td>
<td>0.45</td>
<td>0-2 + 2-4 + 8-16</td>
<td></td>
<td></td>
</tr>
<tr>
<td>E</td>
<td>I</td>
<td>0.45</td>
<td>0-2 + 2-5 + 8-16</td>
<td>~350</td>
<td>~228</td>
</tr>
<tr>
<td>F</td>
<td>I</td>
<td>0.45</td>
<td>0-8 + 8-16</td>
<td></td>
<td></td>
</tr>
<tr>
<td>G</td>
<td>I</td>
<td>0.45</td>
<td>0-8 + 8-16</td>
<td></td>
<td></td>
</tr>
<tr>
<td>H</td>
<td>I</td>
<td>0.45</td>
<td>0-8 + 8-16</td>
<td></td>
<td></td>
</tr>
<tr>
<td>A</td>
<td>II</td>
<td>0.65</td>
<td>0-8 + 8-16</td>
<td>~350</td>
<td>~158</td>
</tr>
<tr>
<td>B</td>
<td>II</td>
<td>0.65</td>
<td>0-18</td>
<td></td>
<td></td>
</tr>
<tr>
<td>C</td>
<td>II</td>
<td>0.65</td>
<td>0-8 + 8-16</td>
<td></td>
<td></td>
</tr>
<tr>
<td>D</td>
<td>II</td>
<td>0.65</td>
<td>0-8 + 8-16</td>
<td></td>
<td></td>
</tr>
<tr>
<td>E</td>
<td>II</td>
<td>0.65</td>
<td>0-8 + 8-16</td>
<td></td>
<td></td>
</tr>
<tr>
<td>A</td>
<td>III</td>
<td>0.45</td>
<td>0-8 + 8-16</td>
<td>~350</td>
<td>~158</td>
</tr>
<tr>
<td>B</td>
<td>III</td>
<td>0.65</td>
<td>0-8 + 8-16</td>
<td></td>
<td>~228</td>
</tr>
</tbody>
</table>

5.3 Methodology according to ISO 11665-7

The International Standardization Organization (ISO) describes a methodology, ISO-11665-7 (2012), which acknowledges two different ways of calculating the exhalation rate of radon gas. One way is an exponential method, where back diffusion (Abo-Elmagd, 2014) is accounted for.
The other approach is according to linear regression analysis. The latter method is proposed as the most usable by ISO and appreciated, due to its much more constrained time frame of measurement (24 hours) and slightly simplified mode of calculation than for the former, simply since back diffusion of $^{222}\text{Rn}$ could be neglected.

Initially, some experiments between the different proposed methodologies to calculate the exhalation rate were conducted on some of the concrete samples. In most cases very similar results were obtained ($\pm 3\%$). However, for some strongly radiating samples, deviations of more than $10\%$ could occur. However, due to the project’s amounts of samples and the multiple analyses at different RH for each sample, the only reasonable option was to use the suggested linear interpolation method.

5.4 Test methods used to calculate the radon gas exhalation rate of a surface

Through linear regression analysis an approximation of the radon gas exhalation ($\text{Bq/m}^2\text{h}$) can be calculated knowing the initial conditions of the radon gas concentration in a given space (volume). Measurements are performed at least for 24h in order to reduce uncertainties. A measurement is taken every 10 minutes yielding a total of at least $(6 \times 24 \text{ h}) = 144$ measurements as the basis for the analysis. The linear model assumes no back diffusion of radon and neglects the decay of radon during time. The equation for the linear regression model can be described as:

$$\phi = \frac{\left(C-C_0\right) \times V}{A}$$

(5.2)

where: $\phi = \text{Exhalation of radon gas (Bq/m}^2\text{h)}$, $C = \text{Concentration of radon gas measured by the radon gas monitor (Bq/m}^3\text{)}$, $C_0 = \text{Background concentration of radon gas at initiation (Bq/m}^3\text{)}$, $t = \text{time of duration (h)}$, $A = \text{Effective surface area of the sample (m}^2\text{)}$, $V = \text{Volume of the container including hoses}$. 

The principal to calculate the exhalation rate is to use a closed system explained in section B of ISO-11665-7. Knowing the surface area of the material, and the volume of the air enclosed (includes volume of container and volume in hoses to connect to the radon-monitor) as well as the background of radon gas at initiation of the measurement ($C_0$), then the necessary variables are known. Figure 5.2 shows the principle of the method.

![Figure 5.2 Principle to evaluate the radon exhalation rate using a closed system. Figure redrawn and modified from ISO 11665-7.](image)

The sample is sealed on each side using a thick butylene-rubber (sticky) with an aluminum-tape as a cover to ensure that no radon gas escapes on the sides or from the bottom of the sample. Figure 5.3
shows the container and the radon gas monitor, when (a) open with sample inserted and (b) closed and sealed with aluminum-tape and clams.

Figure 5.3a Aluminum container with one sample inside resting on the bottom of the container.  
Figure 5.3b Aluminum container in a “sealed state” with clamps and aluminum-tape. The hoses can be seen that are connected to the radon monitor (at the left) creating a “closed system”.

The principal to evaluate the radon gas content stems from an article by Baltzer et al. (1991), where the different photo peaks in keV of $^{222}$Rn, $^{218}$Po, $^{214}$Bi could be well detected, knowing their specific energy. The energy intervals are 6.0, 5.5 and 4.8 for $^{222}$Rn, $^{218}$Po and $^{214}$Bi, respectively. It is thus an indirect method, making use of the proportionality of alpha-decay and its photo peaks of $^{222}$Rn and $^{218}$Po, $^{214}$Po and $^{214}$Bi in relation to content of radon gas within the air.

The microprocessor within the radon monitor subtracts the $^{214}$Po and $^{214}$Bi photo peaks before recalculating the photo peak amplitude of $^{222}$Rn and $^{218}$Po and its relation to content of radon gas (Bq/m$^3$) in air, which is the unit displayed in the window of the radon gas monitor.

A yearly correction factor to the instrument is then applied, after calibration at the Swedish Radiation Safety Authority (SSM) where a well-defined level of $^{222}$Rn is maintained in a given room. The correction factor for the instrument is also used in the final calculation of the exhalation rate using linear regression analysis.

### 5.5 Measurements of Relative Humidity (RH)

The relative humidity is measured by use of the instrument Vaisala HM 44 using calibration equipment HMK 15 to ensure limited drifting of the probes. A yearly calibration has been performed at Vaisala Oy, Helsinki, Finland. The uncertainties, compliant with a confidence level of 95 % result in uncertainty of RH ±1.4.

To maintain a good certainty and assure no drifting of the probes, their initial calibration were checked and if necessary corrected at CBI with the salt-solutions, MgCl$_2$ and LiCl using calibration tables by Greenspan (1977). An uncertainty of ~1.3 % was calculated using these calibration tables at 23°C. In Figure 5.4a, b one of the probes used is presented as well as an ongoing measurement with the Vaisala HM44 equipment.
5.6 Calculation of radon gas (Bq/m$^3$) and influence of ventilation rates

The radon gas content in air (Bq/m$^3$) is calculated applying different ventilation rates within a building. Regulations at hand stipulated by Boverket (National Board of Housing, Building and Planning) and its document *BBR 12* (Swedish National Board of Housing, Building and Planning, 2006) have prescribed a minimum rate of ejected air (ventilation) as 0.35 l/s/m$^2$ across the floor, when people are present within the room/dwelling. Thus, no restriction is made in height, which will inflict on the total volume of the room. As an example a “normal” house, with a floor surface of 10×10 meter in surface area, with a height of 2.4 m would constitute 240 000 l of air in the house.

Volume ($V$) = Height × Width × Length

Recalculations of the flow of 0.35 l/s/m$^2$ to l/h/m$^2$ generate 1260 l/h/m$^2$ in air rate exchange. This means over the full 100 m$^2$ an air rate exchange of 126 000 l/h is achieved.

Hence, for a large living room, 10 m × 10 m in length and width and 2.4 m in height, the living room constitutes 240 000 litres of air (240 m$^3$). This would consequently result in an air rate exchange of 126 000 l/240 000 l = 0.52. Accordingly, a ventilation rate of ~0.5 circulations/h in a normal case.

However, since the regulations in the BBR are related to l/s/m$^2$ surface, the conditions vary depending on the shape of a house/room or dwelling. In a half open space house (often top floor is semi-open), where the height may be doubled (4.8 m) and the surface area (floor) remains 100 m$^2$ at the bottom and an increase by 50 m$^2$ at the top floor the following conditions would apply.
• Volume \((V) = 10 \times 10 \times 4.8\ m = 480\ m^3 = 480\ 000\ liters.\)
• The surface area are of floor \(= 150\ m^2 \times 1260\ l/h/m^2 = 189\ 000\ l/h\) in air exchange rate.
• Thus \(189\ 000/480\ 000 = 0.39\ \text{circ/h}\)

The height (in this case doubled due to two floors, \(2.4\ m \times 2\)) hence has a great impact on the air exchange rate. If we may use the suggested room model as suggested by the CE (2014) similar numbers would apply, where the volume would be,

• Volume in room \(= 3 \times 4 \times 2.5\ m = 30\ m^3\ (30\ 000\ liters)\)
• Room surface area \(= 3 \times 4 = 12\ m^2.\)
• Surface area flow/h \(= 12\ m^2 \times 1260\ \text{liters}/h = 15000\ \text{liters}/m^3 h^{-1}\)
• Yielding a circulation of air in a room of \(0.5\ \text{circ/h}\) (15 000 l/30 000 l)

As explained in the examples, the difference in layouts of houses and its influence on air exchange rate could be very stretchy. Hence, the Thesis presents two different scenarios for the radon gas contribution in air to the room or house. One is a scenario, where “0.3 circ/h” in ventilation rate is used in the calculation and one where “0.5 circ/h” is used.

5.6.1 Phase I

Measurements were done on a regular basis, but in practice, this means that measurements for each concrete sample have been executed with a difference of approximately 10 % in relative humidity (RH). The used w/c ratio was 0.45 for every concrete sample.

The first phase included eight different concrete samples. The concept included simultaneous conservation of an identical “daughter” concrete sample joining each measured “parent” concrete sample. The “parent” and “daughter” concrete samples originated from the same concrete casting specimen, and where sawn in identical proportions as explained in Section 4.4 (Process of sampling). All measurements of RH were performed on the "daughter" concrete sample due to the idea of keeping the surface of the "parent" concrete sample as intact as possible. The concept likewise embraces an identical preservation of the concrete samples if the relative humidity is to be trustworthy. Between measurements and during measurements all concrete samples have been stored in 23°C and 50 RH inside a climate room. Before and during measurements the samples were conditioned in the “measurement room” that has identical conditions in temperature and RH as the climate room. The results are tabulated and presented as graphs in relation to the newly presented model room described by the European Commission in Draft TR 00351020 Dose assessment of emitted gamma radiation for consultation (2014), where the room model is \(3 \times 4 \times 2.5\ m\), as well as the conditions presented by the EC (1999), RP 112, where the room model is \(4 \times 5 \times 2.8\ m\).

5.6.2 Phase II

A scheme to ensure measurement intervals with an approximate 5 % difference in relative humidity (RH) was employed. Measurements of RH were done approximately 1-2 hours before commencement of radon gas measurements. The used w/c ratio was 0.65 for every concrete sample. Five different concrete samples were investigated. The concept included casting of five new concrete samples, each \(300 \times 300 \times 150\ mm\) in size.

The concept also included drilling and insertion of two small plastic tubes 7-8 cm, in depth within the concrete samples. The drilling and insertion of tubes of the samples were done three days after molding. The measurements of RH were done approximately two hours before the initiation of measurement of the radon exhalation rate within the aluminum container.

As part of the study the weight (kg) of each concrete cube was also registered before every new measurement was commenced.
5.6.3 Phase III

A continuation and verification scheme was launched as a consequence of some doubtful preliminary results of phase I at the second half of the project. One concrete sample with a w/c ratio of 0.45 as well as one concrete sample with a w/c ratio of 0.65 were cast using the same aggregate mix. The aggregate mix was identical to one used in one of the concrete portions of phase II. Initial insertion of plastic tubes was done closely after molding (24 hours after mixing the concrete). Weight measurements, relative humidity and the first radon gas exhalation measurements were performed directly thereafter. In order to limit escape of radon gas between the outside of the plastic tubes and the concrete an epoxy-based sealant was used.

As part of this study, the weight (kg) of each concrete cube was also registered before every new measurement was commenced. Measurement of the concretes compaction strength (SS-EN 12390-3) after 7 as well as 28 days was recorded. A mean value of two tested concrete samples for each w/c ratio was examined.
6 Results on gamma radiation measurements

6.1 Assessed materials

The initial number of concrete specimens included ten different raw materials (Table 4.5) where initial results were presented in Papers I and II (Table 6.1). The interested readers will note that there is a difference between the I-indices of the analyzed concrete specimens (portions) presented in Paper III and the results of the concretes presented in Papers I and II. This is due to repeatability measurements of the concrete samples ongoing as Paper I and II were published. Mean values of the I-index of each concrete specimen (portions) have been presented in Paper III (Table 6.2). But the reader should also be aware that these first results are found in the third Paper (Table 2). Hence it should be noted, that in Table 6.3, two columns of analyzed values from STUK are presented. Also, as a consequence the values of concrete as well as I-index in Table 6.2 and 6.3 are mean values.

Table 6.1 The results of the analyzed field spectrometry values (mean) as ambient dose rate (µSv/h). The specific activities (Bq/kg) of $^{40}$K, $^{226}$Ra and $^{232}$Th are shown for ten different concrete specimens with a thickness of 150 mm. Also, the I-index of each concrete measured is calculated from the specific activities of the radionuclides.

<table>
<thead>
<tr>
<th>Concrete sample</th>
<th>Ambient dose rate, $H^o(10)$ (µSv/h)</th>
<th>Specific activity (Bq/kg) - CBI field spectrometry</th>
<th>I-index</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>$^{40}$K</td>
<td>$^{226}$Ra</td>
</tr>
<tr>
<td>1</td>
<td>0.25</td>
<td>1134.63</td>
<td>157.77</td>
</tr>
<tr>
<td>2</td>
<td>0.25</td>
<td>1216.23</td>
<td>92.27</td>
</tr>
<tr>
<td>3</td>
<td>0.20</td>
<td>1283.30</td>
<td>118.70</td>
</tr>
<tr>
<td>4</td>
<td>0.14</td>
<td>748.07</td>
<td>53.72</td>
</tr>
<tr>
<td>5</td>
<td>0.10</td>
<td>983.34</td>
<td>26.14</td>
</tr>
<tr>
<td>6</td>
<td>0.10</td>
<td>871.18</td>
<td>42.71</td>
</tr>
<tr>
<td>7</td>
<td>0.10</td>
<td>852.32</td>
<td>45.13</td>
</tr>
<tr>
<td>8</td>
<td>0.08</td>
<td>704.25</td>
<td>42.48</td>
</tr>
<tr>
<td>9</td>
<td>0.08</td>
<td>606.08</td>
<td>39.63</td>
</tr>
<tr>
<td>10</td>
<td>0.04</td>
<td>381.90</td>
<td>22.00</td>
</tr>
</tbody>
</table>

Later on in the project two more materials were assessed, which were also presented as part of the study in Paper III, including three more materials with a thickness of 200 mm. Table 6.2 presents the results of the gamma spectrometric analysis performed at STUK also including analysis of the aggregates constituting the major part of the concrete. The results were further assessed using a dose calculation by Hoffman (2014). The concept of the dose-calculation by Hoffman (2014) is presented in Paper III (2016).
Table 6.2. The concrete portions analyzed using gamma spectrometric analysis. The difference between aggregates and the final concrete as well as a dose calculation by Hoffman (2014) are included.

<table>
<thead>
<tr>
<th>ID</th>
<th>Activity concentrations of aggregates used in concrete (Bq/kg)</th>
<th>Activity concentrations of concretes (Bq/kg)</th>
<th>Difference between aggregates and concrete in percent (%) for each nuclide</th>
<th>I-index (agg)</th>
<th>I-index (con)</th>
<th>Diff. (Δ) in I-index (%)</th>
<th>Dose-calculation including density correction in brackets</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>(^{40}\text{K})</td>
<td>(^{226}\text{Ra})</td>
<td>(^{232}\text{Th})</td>
<td>(^{40}\text{K})</td>
<td>(^{226}\text{Ra})</td>
<td>(^{232}\text{Th})</td>
<td>(^{40}\text{K})</td>
</tr>
<tr>
<td>1</td>
<td>1371</td>
<td>270.2</td>
<td>192.9</td>
<td>1110</td>
<td>216.7</td>
<td>153.3</td>
<td>19.0</td>
</tr>
<tr>
<td>2</td>
<td>1490</td>
<td>130.0</td>
<td>250.0</td>
<td>1155</td>
<td>96.5</td>
<td>195.0</td>
<td>22.5</td>
</tr>
<tr>
<td>3</td>
<td>1740</td>
<td>180.0</td>
<td>120.0</td>
<td>1445</td>
<td>140.0</td>
<td>99.5</td>
<td>17.0</td>
</tr>
<tr>
<td>3 (200)</td>
<td>1740</td>
<td>180.0</td>
<td>120.0</td>
<td>1410</td>
<td>140.0</td>
<td>98.0</td>
<td>19.0</td>
</tr>
<tr>
<td>4</td>
<td>770</td>
<td>40.5</td>
<td>30.8</td>
<td>730</td>
<td>52.7</td>
<td>102.3</td>
<td>5.2</td>
</tr>
<tr>
<td>4 (200)</td>
<td>770</td>
<td>40.5</td>
<td>30.8</td>
<td>690</td>
<td>47.0</td>
<td>95.5</td>
<td>10.4</td>
</tr>
<tr>
<td>5</td>
<td>1160</td>
<td>43.0</td>
<td>54.0</td>
<td>905</td>
<td>38.0</td>
<td>42.5</td>
<td>22.0</td>
</tr>
<tr>
<td>6</td>
<td>1115</td>
<td>50.2</td>
<td>52.2</td>
<td>867</td>
<td>38.0</td>
<td>42.0</td>
<td>22.2</td>
</tr>
<tr>
<td>7</td>
<td>1060</td>
<td>19.0</td>
<td>44.0</td>
<td>1045</td>
<td>19.7</td>
<td>41.0</td>
<td>1.4</td>
</tr>
<tr>
<td>7B</td>
<td>1060</td>
<td>19.0</td>
<td>44.0</td>
<td>1010</td>
<td>16.3</td>
<td>41.5</td>
<td>4.7</td>
</tr>
<tr>
<td>8</td>
<td>817</td>
<td>43.9</td>
<td>33.1</td>
<td>650</td>
<td>33.4</td>
<td>27.6</td>
<td>20.5</td>
</tr>
<tr>
<td>9</td>
<td>620</td>
<td>44.0</td>
<td>25.0</td>
<td>539</td>
<td>32.8</td>
<td>25.9</td>
<td>13.1</td>
</tr>
<tr>
<td>10</td>
<td>287</td>
<td>7.11</td>
<td>6.5</td>
<td>261</td>
<td>8.2</td>
<td>7.3</td>
<td>9.1</td>
</tr>
<tr>
<td>11</td>
<td>470</td>
<td>21.0</td>
<td>21.0</td>
<td>390</td>
<td>17.9</td>
<td>16.2</td>
<td>17.0</td>
</tr>
<tr>
<td>12</td>
<td>1180</td>
<td>121.5</td>
<td>69.0</td>
<td>925</td>
<td>104.7</td>
<td>70.0</td>
<td>21.6</td>
</tr>
<tr>
<td>12 (200)</td>
<td>1180</td>
<td>121.5</td>
<td>69.0</td>
<td>955</td>
<td>115.0</td>
<td>73.0</td>
<td>19.1</td>
</tr>
</tbody>
</table>

The difference (Δ) in I-index (%) is calculated as “[I-index (agg)] – [I-index (con)]) / [I-index (agg)]”. Noticeably the difference (Δ) in I-index is for almost every sample neutral or positive and varies from zero up to ~23%. But, there is a large discrepancy for ID-sample 4, marked with italics, with negative values. The large negative numbers for sample ID “4” are most likely due to incorrect sampling. This discrepancy is further discussed in Chapter 8 – “Discussion”. The overall positive values highlights that the final concrete in relation to the aggregate used in general has a lower I-index. Accordingly it also indicates a dilution effect of the cement containing a limited abundance of the radioactive isotopes \(^{40}\text{K}, \, ^{226}\text{Ra}, \, ^{232}\text{Th}\).

In Figure 6.1 the results of the presented I-indices in Table 6.1 of the concrete specimens are presented excluding the samples with a thickness of 200 mm, no 3 (200) and 4 (200), as well as 7B (w/c ratio 0.65) and no 12 of Table 6.2. A blind test is done for sample 11, shown as sample 12 in the figure. A comparison with geochemical analysis of the same concrete portions at CRPG-CNRS was assessed simultaneously.
Figure 6.1 Comparative data using the I-index screening tool model for field spectrometry as well as laboratory gamma spectrometry as geochemical analysis of 150 mm thick concretes.

A more thorough overview of the comparative data are presented in Table 6.3. All in all, eleven different concrete samples (excluding on duplicate) have been sent for geochemical analysis and compared as shown in Figure 6.1.
Table 6.3 Comparison between analytical results from CBI, STUK and CRPG/CNRS using gamma ray spectrometry and geochemical analysis (ICP-MS/EOS). The calculated I-indices related to the measurements of each analysis are compared as ratios. The initially calculated I-indices due to first assessment of results from STUK are also included. In italics (sample 8) not part of Papers I and II.

<table>
<thead>
<tr>
<th>Samples (concrete)</th>
<th>I-indices</th>
<th>Ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>CBI</td>
<td>STUK(^5)</td>
</tr>
<tr>
<td>1</td>
<td>0.28</td>
<td>0.15</td>
</tr>
<tr>
<td>2</td>
<td>0.49</td>
<td>0.44</td>
</tr>
<tr>
<td>3</td>
<td>0.53</td>
<td>0.50</td>
</tr>
<tr>
<td>4</td>
<td>0.64</td>
<td>0.59</td>
</tr>
<tr>
<td>5</td>
<td>0.65</td>
<td>0.64</td>
</tr>
<tr>
<td>6</td>
<td>0.65</td>
<td>0.65</td>
</tr>
<tr>
<td>7</td>
<td>0.89</td>
<td>1.02</td>
</tr>
<tr>
<td>8</td>
<td>0.95</td>
<td>0.78</td>
</tr>
<tr>
<td>9</td>
<td>1.26</td>
<td>1.49</td>
</tr>
<tr>
<td>10</td>
<td>1.56</td>
<td>1.79</td>
</tr>
<tr>
<td>11</td>
<td>1.58</td>
<td>2.04</td>
</tr>
<tr>
<td></td>
<td>Average</td>
<td>1.09</td>
</tr>
<tr>
<td></td>
<td>Median</td>
<td>1.05</td>
</tr>
</tbody>
</table>

At lower I-index values ~0.5 (see Figure 6.1) all CBI measurements yield slightly higher results than results obtained from STUK and CRPG/CNRS. Secondly, for I-index > 1 a trend could be seen that the analytical measurements performed by STUK or CRPG/CNRS converted to I-index are increasingly greater than measurements performed by CBI. Around values of I-Index ~1 and below, the correlation between different analytical practices show a good to moderate agreement (< 10 % difference). The analytical methodologies show overall a better correlation (Table 6.3) using the ratio to compare the methods.

Field measurements were also undertaken in order to compare the analytical gamma ray spectrometry methodology with the laboratory gamma spectrometer techniques used by STUK. A special focus was drawn to higher values as shown in Figure 6.1. Bedrock samples were taken from quarries, where flat surfaces were recognized (having 2\(\pi\) geometry) that could be measured by the hand-held spectrometer. Table 6.4 presents comparative data between the I-indices calculated from field measurements of the elemental concentrations and recalculated to specific activity (Bq/kg) of the radionuclides compared to when loose rock was sampled from the measurement site and sent for laboratory gamma spectrometric analysis. In Table 6.4 a reference under the “ID-column” is also made to the “ID-number” in Paper III marked as superscript. The reference of the bedrock material could also be found in Table 4.5.

\(^5\) First assessment of results yielded I-indices as presented in Papers I and II.
Table 6.4 Comparative data of natural radioactivity of four different rock types (a), firstly measured in the field (b) and thereafter, sampled loose rock at the measurement site was sent for gamma spectrometric evaluation (c). Data presented as I-index. A mean value of 0.93 (d) was calculated for the four samples. In the column “ID” a notation with superscript is made as to address the correct “ID” as presented in Paper III.

<table>
<thead>
<tr>
<th>ID</th>
<th>Samples (rock)</th>
<th>Field measurement, rock (I-index)</th>
<th>STUK, rock sent for analysis (I-index)</th>
<th>Ratio (b)/(c)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1&lt;sup&gt;a&lt;/sup&gt;</td>
<td>Granitoid</td>
<td>2.05</td>
<td>2.21</td>
<td>0.92</td>
</tr>
<tr>
<td>2&lt;sup&gt;b&lt;/sup&gt;</td>
<td>Granitoid</td>
<td>2.10</td>
<td>2.11</td>
<td>1.00</td>
</tr>
<tr>
<td>3&lt;sup&gt;c&lt;/sup&gt;</td>
<td>Gneiss</td>
<td>0.66</td>
<td>0.80</td>
<td>0.82</td>
</tr>
<tr>
<td>4&lt;sup&gt;a&lt;/sup&gt;</td>
<td>Granitoid</td>
<td>0.48</td>
<td>0.48</td>
<td>1.00</td>
</tr>
</tbody>
</table>

6.2 Measurements of crushed aggregates and concrete – Ambient dose equivalent rate H*(10)

The different particle size fractions were measured by use of the portable hand-held gamma ray spectrometer, when the material arrived at CBI, Borås. Measurements were performed when the material was slightly dried during sunny weather conditions. Figure 6.2 presents the difference between the finalized concrete specimen (cement, water, aggregates and additives) and the sole aggregates. From the results it is apparent that some aggregates contribute with high elemental concentrations of the natural radioisotopes. Secondly, the cement, water and additives are all low in their elemental concentrations of the natural radionuclides (\(^{40}\)K, \(^{226}\)Ra, \(^{232}\)Th). Hence, the results at higher values of the concrete mixes could be understood as an obvious dilution of the specific activity (Bq/kg) of the aggregates. This in turn yields a marked change in the assessed ambient dose rate (\(\mu\)Sv/h) of the concrete specimens. Aggregates are most often the constituent that contributes with excessive amounts of radionuclides in the building material.

One sample has been produced by using mixed aggregates, such as glaciofluvial sediments (A) and crushed bedrock (AG). Therefore, this material has a letter “A” for particle size fraction 0-8 mm and “GA” for particle size fraction 8-16 mm. The outcome, containing the mixed aggregates in the concrete cast is shown with letter ”C”.

53
Figure 6.2 The ambient dose equivalent rate (µSv/h) for the crushed materials and the concrete mixes containing the same aggregate. The letter “A” and “C” denote crushed aggregate and concrete, respectively. The red dotted line is in order to illustrate the reference value of 0.3 µSv/h in gamma radiation legislated by the Swedish National Board of Housing, Building and Planning (2011).

The reference value (0.3 µSv/h) is in relation to earlier recommendations put forward by the Swedish Radiation Safety Authority, which currently are implemented in the regulatory standard published by the National Board of Housing, Building and Planning, BBR 12 (2011).

From the presented results, the initial differences are obvious between aggregates and concrete, but for lower values, the ambient dose rate ~< 0.1 µSv/h, the dilution effect between the crushed aggregate, and when this aggregate is part of the concrete is almost vanishing.

6.3 Measurements of crushed aggregates and concrete - Activity Index (I)

Figure 6.3 shows the same assessed materials as in Figure 6.2, but presented when recalculated according to the I-index. Consequently they could be assessed in line with the recommendations of the European Councils directive of Basis Safety Standards released in January 2014 (CE, 2014).
Figure 6.3 The I-index as a function of crushed materials (aggregates) and when the aggregates constitute part of the concrete. The letters “A” and “C” denote crushed aggregate and concrete, respectively. The red dotted line is presented in order to illustrate the screening tool value of 1 in I-index.

6.4 Link between I-index and dose model (effective dose)

In this Thesis the analytical results of the gamma spectrometry analysis were used to investigate the difference between the I-index (EC, 1999) and a dose model (Hoffman, 2014), that are put forward by the European Commission in their forthcoming technical report presented to the WG 3 in July 2016.

The results are more thoroughly presented in Paper III, but are shown in Figure 6.4. The Figure presents two scenarios, where sample “ID nr 4” as shown in Table 6.2 has received analytical results, which are very dubious. Thus the two regression lines presented in Figure 6.4 displays two scenarios, where the ID-number analyse is excluded and the other option when included. The results are to be viewed in the light of the recipe used by CBI (Section 6.7, Table 6.5).
Figure 6.4. The effective dose of concretes as a function of the I-index of aggregates using a dose model by Hoffman (2014) to calculate the effective dose of the concretes. Two regression lines, where ID-sample 4 is included in one analysis and excluded in the other one. The two blue squares lacking uncertainty bars are due to this approach.

The correlation ($R^2$) is strong and yields an approximate relation of an effective dose of 0.65 mSv per year from concrete in excess dose of the building materials as the I-index of the aggregates is equal to 1. The above conditions are in relation to the draft, WI 00351020 Dose assessment of emitted gamma radiation for consultation (2014), where the room model is $3 \times 4 \times 2.5$ m with 200 mm thick concrete walls.

6.5 Links between exposure (X), absorbed dose (D), ambient equivalent dose ($H^{*}10$) and I-index

The instrument RS 230 uses conversion factors between exposure (X), absorbed dose in air (D) and absorbed dose for soft tissue as to get the quantities and their respective units in ambient dose equivalent rate ($\mu$Sv/h), absorbed dose rate in air (nGy/h) and röntgen ($\mu$R/h) earlier mentioned in Subsection 4.2.1.

The instrument is calibrated with the correlation that, the exposure rate of 0.996 $\mu$R $h^{-1}$ ($\sim 1 \mu$Rh$^{-1}$), which applies to air as the irradiated medium and to gamma rays as the radiation, results in an absorbed dose rate in soft tissue of $\sim 1 \times 10^{-2}$ $\mu$Gy $h^{-1}$ (10 nGy $h^{-1}$). An exposure of $\sim 1 \mu$R in air, over a rather wide range of photon energies yields an absorbed dose in air of about 8.76 nGy based on International Commission on Radiation Units and Measurements (ICRU, 1998) conversion factors. IAEA (2003) relates to a conversion factor of 8.69 nGy/$\mu$R in air.

The absorbed dose in air can then be converted to absorbed dose in soft tissue by assuming that secondary charged particle equilibrium exists in the irradiated tissue and multiplying the air absorbed dose by the ratio of the mass energy absorption coefficient for soft tissue compared to air (Isaksson, 2011). This ratio changes slowly over a wide photon-energy interval, and a value of $\sim 1.1$ may be fairly representative (IAEA, 2003, ICRP, 2012) at energies around 1 MeV. Multiplying the absorbed dose of 8.76 nGy in air by this factor yields an absorbed dose in soft tissue $\sim 9.6$ nGy ($\sim 1 \times 10^{-2}$ $\mu$Gy), which means that 1 $\mu$Rh$^{-1}$ $\sim 0.1$ $\mu$Sv/h ($\sim 1 \times 10^{-2}$ $\mu$Gy) ambient dose equivalent rate ($H^{*}10$).
A plot to describe the relation between the RS 230’s measured ambient dose rate (H*10) and the calculated specific activities of the natural radioisotopes, displayed as I-index is shown in Figure 6.5.

![Graph](image)

**Figure 6.5.** The calculated I-index of the concrete specimens specific activities (Bq/kg) as a function of the ambient equivalent dose rate, H*10 in µSv/h measured by the gamma hand held spectrometer. A linear regression analysis (dotted line) is calculated and represented in the Figure.

The approximate linear correlation between the ambient equivalent dose rate H'10 and the I-index implies that the instrument is correctly tuned between specific activity (Bq/kg) presented as I-index and its measured ambient equivalent dose rate (H*10).
6.6 CBI methodology - Results in relation to I-index

Figure 6.6 I-index of the small concrete slabs (300 × 300 × 150) mm as a function of I-index of the large concrete specimens (1500 × 1500 × 150) mm.

Figure 6.6 describes the relation (ratio) between the large concrete specimens and the small concrete samples measured. The dashed dotted line describes the linear relation that could be used to estimate I-index. The relation described gives a specific relation between the geometries of the different concrete sizes using an identical recipe. Hence, the linear correlation is not 1 to 1, which would be the case if measurements were performed on two equal concrete specimens containing the same building materials (constituents). As shown in Figure 6.6, the k-value according to the linear regression analysis is ~0.5. As a consequence, one realize, that the small concrete sample only registers approximately 50 % of the natural radioisotopes in relation to its “true value” as when measurements are performed on the larger concrete specimens.

By making use of regression analysis and the R²-value, we could get a good idea of the reproducibility of the results (Garcia, 2010). An R-value ~1 means that the y-variable has a strong dependence of the independent variable (x), Garcia (2010). In this case, by use of a correlation coefficient we could estimate the approximate value of either the total dose rate/h or the I-index for the given concrete mix.

Comparative measurements have been done for 150 and 200 mm thick concrete mixes as well. The results are illustrated in Figure 6.7
6.7 Calculation of the I-index of building materials according to BSS

In Subsection 6.7.1 a calculation of the I-index of a concrete constituted of several constituents is exemplified. The theoretical approach is in line with the forthcoming European harmonized standard (TS 00351014) and the suggested approach in line with article 75 within the EU’s BSS (EC, 2014).

In Subsection 6.7.2 there are some comparative results, when the examined concrete specimens are theoretically calculated knowing the activity (Bq/kg) of each constituent and compared to the analyzed results of the concretes samples sent to STUK. In Subsection 4.3.3 a general recipe of the cast concrete is also presented.

6.7.1 A model for calculation

In the following example (Table 6.4) a calculation of a simple concrete mix is demonstrated. The following components are included;

1. Water
2. Cement
3. Aggregates, crushed bedrock (particle size fraction 8-16 mm)
4. Aggregates, fluvial sediments (particle size fraction 0-8 mm)

Additives, such as plasticizers or air-entrainment agents are in the example neglected.

The concentrations of radionuclides are calculated as decays/s/kg (Bq/kg) and consequently the mass of the material is in focus. Calculating the volume necessary for the production of 1 m³ of concrete, the density and mass of each material has to be known variables. The bulk density of the constituents is the correct density to be used.
Table 6.5. Mix design of a concrete recipe with w/c ratio 0.45 and air content set to 1 %.

<table>
<thead>
<tr>
<th>Recipe</th>
<th>Volume (m³)</th>
<th>Mass (kg)</th>
<th>Weight (%)</th>
<th>Density (kg/m³)</th>
<th>Abbreviations for mass</th>
</tr>
</thead>
<tbody>
<tr>
<td>Water</td>
<td>0.158</td>
<td>158</td>
<td>6.5</td>
<td>1000</td>
<td>( m_w )</td>
</tr>
<tr>
<td>Cement</td>
<td>0.114</td>
<td>350</td>
<td>14.5</td>
<td>3080</td>
<td>( m_c )</td>
</tr>
<tr>
<td>Aggregates (0/8)</td>
<td>0.345</td>
<td>914</td>
<td>37.9</td>
<td>2650</td>
<td>( m_{A,0/8} )</td>
</tr>
<tr>
<td>Aggregates (8/16)</td>
<td>0.374</td>
<td>991</td>
<td>41.1</td>
<td>2650</td>
<td>( m_{A,8/16} )</td>
</tr>
<tr>
<td>Air (1 %)</td>
<td>0.01</td>
<td>0.012</td>
<td>-</td>
<td>1.2</td>
<td>-</td>
</tr>
<tr>
<td><strong>Total</strong></td>
<td>1</td>
<td>2413</td>
<td></td>
<td></td>
<td>( m_t )</td>
</tr>
</tbody>
</table>

Each constituent’s natural radioactivity also has to be known. After evaluation of each constituent’s activity concentration of \(^{40}\text{K}, \, ^{226}\text{Ra}\) and \(^{232}\text{Th}\) in decays/s/kg or Bq/kg by use of gamma spectrometric analysis, all the necessary data are at hand.

The calculation of the final activity concentration in Bq/kg for each nuclide in the finalized concrete product could be summarized as:

\[
AC = \sum AC_w \times \left( \frac{m_w}{m_t} \right) + AC_c \times \left( \frac{m_c}{m_t} \right) + AC_{A\,0/8} \times \left( \frac{m_{A\,0/8}}{m_t} \right) + AC_{A\,8/16} \times \left( \frac{m_{A\,8/16}}{m_t} \right)
\]

(6.1)

where, \( AC \) = activity concentration of each nuclide, \( AC_w \) = activity concentration in water, \( AC_c \) = activity concentration in cement, \( AC_{A\,0/8} \) = activity concentration of aggregates 0-8 mm, \( AC_{A\,8/16} \) = activity concentration of aggregates 8-16 mm. \( m_c, m_w, m_{A\,0/8}, m_{A\,8/16} \) are masses (kg) of constituents found in Table 6.5 and \( m_t \) = total mass of 1 m³ of concrete.

To get the total count rate in either nGy/h or µSv/h of absorbed dose rate or ambient dose rate (H\(^*\)10), respectively, of each nuclide use the conversion factors in Table 4.1 to relate activity to parts per million or % and then convert to correct unit by using Table 4.3.

In order to calculate the I-index the following applies, where \( C \) = concentration in Bq/kg (the I-index is dimensionless).

\[
I = \frac{C^{40K}}{3000 \text{ Bq/kg}} + \frac{C^{226Ra}}{300 \text{ Bq/kg}} + \frac{C^{232Th}}{200 \text{ Bq/kg}}
\]

(6.2)

where, \( C^{40K}, C^{226Ra} \) and \( C^{232Th} \) denotes the concentration (“C”) of each radionuclide in Bq/kg.

Adding several components, such as fly ash, fillers or other additives the actual activity (I) of the concrete is calculated in the same manner as described above. The example continues in the next Subsection, 6.7.2.
6.7.2 Comparative results between theoretical calculation and analyzed building material

Each constituent used as part of the concrete has been assessed in relation to its activity. This is also addressed in Paper III as to show the influence of each component. Knowing that the cement and water are fixed in proportion (see Table 6.5) with a set w/c-ratio it is only the input of different quantities of aggregates and their weight (%) that varies in the recipe. Table 6.6 presents the calculated theoretical value of the final specific activity, presented as I-index using the results of the analyzed specific activity (Bq/kg) of each constituent. These values are compared to the results of the analyzed concretes in order to check the quality of the results. The concept of calculating the final activity of the building material as I-index is described by Formula 6.2 (Subsection 6.7.1). The different densities are also related to the calculated density of the recipe compared to the measured density of the building material. Two blind tests were also performed and these ID-numbers are presented in “italics”.

Table 6.6. Theoretically calculation of the final specific activity presented as I-index compared to the analyzed result of the building material (STUK-result). The total mass of 1 m$^3$ of concrete according to the recipe compared to the actual measurements of density using small concrete cubes is also included.

<table>
<thead>
<tr>
<th>ID</th>
<th>Density-recipe (kg/m$^3$)</th>
<th>Density-measured</th>
<th>CBI-calculated</th>
<th>STUK-results</th>
<th>Difference$^6$ (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>I-index</td>
<td>I-index</td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>2396</td>
<td>2288</td>
<td>1.81</td>
<td>1.86</td>
<td>2.4</td>
</tr>
<tr>
<td>2</td>
<td>2382</td>
<td>2260</td>
<td>1.73</td>
<td>1.68</td>
<td>-3.1</td>
</tr>
<tr>
<td>3</td>
<td>2400</td>
<td>2338</td>
<td>1.46</td>
<td>1.45</td>
<td>-0.7</td>
</tr>
<tr>
<td>4</td>
<td>2509</td>
<td>2423</td>
<td>0.47</td>
<td>0.93</td>
<td>49.5</td>
</tr>
<tr>
<td>5</td>
<td>2392</td>
<td>2310</td>
<td>0.66</td>
<td>0.64</td>
<td>-2.4</td>
</tr>
<tr>
<td>6</td>
<td>2390</td>
<td>2359</td>
<td>0.64</td>
<td>0.63</td>
<td>-1.8</td>
</tr>
<tr>
<td>7</td>
<td>2388</td>
<td>2355</td>
<td>0.53</td>
<td>0.62</td>
<td>14.0</td>
</tr>
<tr>
<td>8</td>
<td>2389</td>
<td>2355</td>
<td>0.53</td>
<td>0.60</td>
<td>11.4</td>
</tr>
<tr>
<td>9</td>
<td>2389</td>
<td>2417</td>
<td>0.49</td>
<td>0.47</td>
<td>-4.7</td>
</tr>
<tr>
<td>10</td>
<td>2590</td>
<td>2562</td>
<td>0.30</td>
<td>0.27</td>
<td>-9.1</td>
</tr>
<tr>
<td>11</td>
<td>2404</td>
<td>2418</td>
<td>0.97</td>
<td>1.07</td>
<td>9.1</td>
</tr>
<tr>
<td>11</td>
<td>2404</td>
<td>2418</td>
<td>0.97</td>
<td>1.01</td>
<td>3.7</td>
</tr>
</tbody>
</table>

$^6$ The difference (%) is calculated as ("STUK-results – “CBI calculated “) / “STUK-results") × 100
7 Results – radon gas in a room (Bq/m³)

7.1 Model for calculation of the radon gas within a room

The model applied for calculation of the radon gas content within a room is described by Åkerblom and Clavensjö (2004, 2007). It is a recommended approach by the National Board of Housing, Planning and Building (2011) as a guide to estimate the radon gas content within a room. The following equation applies for calculation of the radon gas content in air within a room, taking the ventilation rate into account:

\[ C_m = \left( \frac{1}{r+n} \right) \times \left( \frac{E \times F}{V} \right) \]  

(7.1)

where, \( C_m \) = Radon gas in air (Bq/m³), \( n \) = air exchange rate (ventilation) in circulations/h, \( r \) = decay constant of radon gas (0.00755), \( E \) = exhalation rate of the material (Bq/m²h), \( F \) = area of exhalation (surface) within the room (m²), \( V \) = Volume of the room (m³).

7.2 Measurements and variables

The figures presented illustrate the humidity of the concrete block as a function of time (years). The humidity is calculated as a mean result of two measurements from the middle of the concrete cubes.

The results are presented with two different ventilation rates. The influence of ventilation rates is discussed in Subsection 5.6. In addition to presenting calculated results of two different rates of circulation within a room, considerations have also been taken to whether only walls are used in the room or if the whole room is built with solid concrete. Consequently, two different scenarios are presented for each ventilation rate;

1. The first scenario with a given circulation/h and only considering the walls made up of concrete. This is in the legend, denoted by "w" for walls.
2. The second scenario considers walls, floor and roof made up of concrete. In the legend any excess symbol for the case of “walls, floor and roof” is left out.

For the presented scenarios, considering a room, the volume of the room and the area are variables of importance. These variables will inflict on the final radon gas contribution (Bq/m³) within the specified room. In the presented scenarios (Table 7.1), the model room dimensions at hand are as suggested by the European commission in RP 112 (1999) and the recently drafted work item -TR 00351020 put forward by the EC in 2014 and anticipated to be finalized in 2017/18.
Table 7.1. Different room dimensions, volume and area taking into consideration the accepted model according to RP 112 (1999) and a suggested room model by the EC (2014) presented in their work item -TR 00351020.

<table>
<thead>
<tr>
<th>Dimensions (m), area (m²) and volume (m³)</th>
<th>RP 112 (1999)</th>
<th>(EC 2014)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Height</td>
<td>2.8</td>
<td>2.5</td>
</tr>
<tr>
<td>Width</td>
<td>4.0</td>
<td>3</td>
</tr>
<tr>
<td>Length</td>
<td>5.0</td>
<td>4</td>
</tr>
<tr>
<td>Total volume in room (m³)</td>
<td>56</td>
<td>30</td>
</tr>
<tr>
<td>Total area, walls (m²)</td>
<td>50.4</td>
<td>35</td>
</tr>
<tr>
<td>Total area, floor and roof (m²)</td>
<td>40</td>
<td>24</td>
</tr>
<tr>
<td>Total area, floor, roof and walls (m²)</td>
<td>90.4</td>
<td>59</td>
</tr>
</tbody>
</table>

The implications of changing room dimensions are shown in Table 7.2, where a moderate to highly radiating concrete has an exhalation rate of 27.7 Bq/m²h. A slight increase in radon gas content (Bq/m³) due to the smaller room size is shown for both a ventilation rate of 0.3 and 0.5.

Table 7.2 Comparison of the change in radon gas contribution by changing the specified conditions of room dimensions.

<table>
<thead>
<tr>
<th>Model</th>
<th>Radon exhalation rate (Bq/m²h)</th>
<th>Circulation/hour in ventilation rate</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Cm (Bq/m³)</td>
<td>0.5</td>
</tr>
<tr>
<td></td>
<td></td>
<td>0.3</td>
</tr>
<tr>
<td>Area included</td>
<td>Only walls</td>
<td>All surfaces</td>
</tr>
<tr>
<td></td>
<td>Only walls</td>
<td>All surfaces</td>
</tr>
<tr>
<td>RP 112 (1999)</td>
<td>27.7</td>
<td>49</td>
</tr>
<tr>
<td></td>
<td></td>
<td>93</td>
</tr>
<tr>
<td></td>
<td></td>
<td>81</td>
</tr>
<tr>
<td></td>
<td></td>
<td>153</td>
</tr>
<tr>
<td>EC (2014)</td>
<td>64</td>
<td>107</td>
</tr>
<tr>
<td></td>
<td></td>
<td>105</td>
</tr>
<tr>
<td></td>
<td></td>
<td>177</td>
</tr>
</tbody>
</table>
7.3 Phase I – 0.45 w/c ratio

7.3.1 General

In the first phase, nine different concrete samples were assessed. In Table 7.3 the data needed as to calculate the final radon gas level within a room is presented in line with the aggregates/bedrock constituting part of the concrete. The relation between the concrete specimens used for gamma spectrometry analysis and the concrete slabs used for radon gas evaluation could be found in Table 4.5.

The calculations of the radon gas in air within a specified room as presented in Table 7.1 and 7.2 are shown in Figures 7.1-7.4 with two different ventilation rates (0.3 and 0.5 circ./h).

Table 7.3 Measured relative humidities (RH %) and calculated exhalation rates of each specific sample as well as used aggregates/bedrock in the concrete.

<table>
<thead>
<tr>
<th>Id</th>
<th>Bedrock</th>
<th>RH (%)</th>
<th>Radon gas exhalation rate, $E$ (Bq/m$^2$h)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sample 1</td>
<td>Granitoid</td>
<td>87</td>
<td>18.3</td>
</tr>
<tr>
<td></td>
<td></td>
<td>78</td>
<td>27.9</td>
</tr>
<tr>
<td></td>
<td></td>
<td>76</td>
<td>27.0</td>
</tr>
<tr>
<td></td>
<td></td>
<td>71</td>
<td>27.7</td>
</tr>
<tr>
<td>Sample 2</td>
<td>Granitoid</td>
<td>84</td>
<td>34.2</td>
</tr>
<tr>
<td></td>
<td></td>
<td>73</td>
<td>36.5</td>
</tr>
<tr>
<td></td>
<td></td>
<td>68</td>
<td>42.6</td>
</tr>
<tr>
<td></td>
<td></td>
<td>60</td>
<td>39.9</td>
</tr>
<tr>
<td></td>
<td></td>
<td>54</td>
<td>39.1</td>
</tr>
<tr>
<td>Sample 3</td>
<td>Gneiss</td>
<td>85</td>
<td>3.8</td>
</tr>
<tr>
<td></td>
<td></td>
<td>67</td>
<td>4.3</td>
</tr>
<tr>
<td>Sample 4</td>
<td>Granitoid</td>
<td>86</td>
<td>4.8</td>
</tr>
<tr>
<td></td>
<td></td>
<td>71</td>
<td>10.3</td>
</tr>
<tr>
<td></td>
<td></td>
<td>67</td>
<td>9.5</td>
</tr>
<tr>
<td>Sample 5</td>
<td>Granitoid</td>
<td>76</td>
<td>66.8</td>
</tr>
<tr>
<td></td>
<td></td>
<td>73</td>
<td>59.2</td>
</tr>
<tr>
<td></td>
<td></td>
<td>68</td>
<td>66.7</td>
</tr>
<tr>
<td></td>
<td></td>
<td>63</td>
<td>76.0</td>
</tr>
<tr>
<td></td>
<td></td>
<td>58</td>
<td>65.8</td>
</tr>
<tr>
<td></td>
<td></td>
<td>53</td>
<td>66.0</td>
</tr>
<tr>
<td>Sample 6</td>
<td>Gneiss + GS</td>
<td>88</td>
<td>6.7</td>
</tr>
<tr>
<td></td>
<td></td>
<td>74.6</td>
<td>6.8</td>
</tr>
<tr>
<td>Sample 7</td>
<td>Granitoid/gneiss</td>
<td>75</td>
<td>27.6</td>
</tr>
<tr>
<td></td>
<td></td>
<td>70</td>
<td>42.6</td>
</tr>
<tr>
<td></td>
<td></td>
<td>63</td>
<td>42.9</td>
</tr>
<tr>
<td>Sample 8</td>
<td>Granitoid/gneiss</td>
<td>90.8</td>
<td>11.6</td>
</tr>
<tr>
<td></td>
<td></td>
<td>77.2</td>
<td>11.3</td>
</tr>
<tr>
<td></td>
<td></td>
<td>X</td>
<td>10.2</td>
</tr>
<tr>
<td>Sample 9</td>
<td>Granitoid/gneiss</td>
<td>81.0</td>
<td>43.1</td>
</tr>
<tr>
<td></td>
<td></td>
<td>69.2</td>
<td>37.8</td>
</tr>
</tbody>
</table>
7.3.2 Radon gas in air within a room having 0.3 or 0.5 circulations/h

Figure 7.1 presents two scenarios where the ventilation rates are calculated as 0.3 and 0.5 circulations/h using the room defined by EC (2014), Table 7.1. The first condition in Figure 7.1 is calculated, whereas only the walls are made of concrete and are the sole contributor to the increase of radon gas within the room. In Figure 7.2 the identical scenario of ventilation rates are presented as in Figure 7.1, but including that every wall, floor and roof are considered to be cast in concrete.

Figure 7.1. Calculated radon gas content in air (Cm) for different concretes as a function of Relative Humidity (RH) where only walls (w) are considered as contributors of the radon gas. Two different ventilations rates are calculated. The dotted lines represent the calculated radon gas rates of the concretes using a ventilation rate of 0.3 circ./h. The solid lines represent a ventilation rate of 0.5 circ./h.

Figure 7.2. Calculated radon gas content in air (Cm) for different concretes as a function of Relative Humidity (RH) where every wall, roof and floor of the room consists of concrete. Two different ventilations rates are calculated. Only a few samples are plotted to demonstrate the influence of an increased ventilation rate. The dotted lines represent the calculated radon gas rates of the concretes using a ventilation rate of 0.3 circ./h. The solid lines represent a ventilation rate of 0.5 circ./h.
7.4 Phase II – 0.65 w/c ratio

7.4.1 General

In the second phase, five concrete samples were assessed. The w/c ratio was set to 0.65. In Table 7.4 the measured relative humidity’s and calculated exhalation rates are presented in conjunction with the link to the bedrock materials used in the first study. The calculations of the radon gas within a room as a function of RH (%) and time (t) are presented in Figures 7.5 and 7.6, respectively, with a the ventilation rate of 0.5 circ./hour.

Table 7.4. Measured and calculated exhalation rates of each specific sample.

<table>
<thead>
<tr>
<th>Id-nummer</th>
<th>Bedrock</th>
<th>Aggregates used - link to phase I samples</th>
<th>RH (%)</th>
<th>Radongas exhalation rate, $E$ (Bq/m²h)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sample 1</td>
<td>Granitoid</td>
<td>Identical aggregate as in sample 5</td>
<td>91.8</td>
<td>108.8</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>84.9</td>
<td>94.3</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>83.2</td>
<td>73.8</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>73.1</td>
<td>62.9</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>66.2</td>
<td>71.8</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>61.0</td>
<td>58.7</td>
</tr>
<tr>
<td>Sample 2</td>
<td>Granitoid/gneiss</td>
<td>Mix of aggregates, sample 5 and sample 8</td>
<td>92.0</td>
<td>70.1</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>87.6</td>
<td>62.5</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>78.2</td>
<td>45.2</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>71.3</td>
<td>50.2</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>64.2</td>
<td>41.4</td>
</tr>
<tr>
<td>Sample 3</td>
<td>Granitoid/gneiss</td>
<td>Identical aggregate as in sample 9</td>
<td>93.5</td>
<td>57.8</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>87.5</td>
<td>44.2</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>80.0</td>
<td>38.4</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>64.0</td>
<td>38.9</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>59.1</td>
<td>32.0</td>
</tr>
<tr>
<td>Sample 4</td>
<td>Granitoid</td>
<td>Identical aggregate as in sample 2</td>
<td>94.0</td>
<td>49.2</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>88.5</td>
<td>36.9</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>81.9</td>
<td>35.6</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>66.5</td>
<td>33.5</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>60.9</td>
<td>28.3</td>
</tr>
<tr>
<td>Sample 5</td>
<td>Granitoid/gneiss</td>
<td>Identical aggregate as in sample 8</td>
<td>96.0</td>
<td>12.8</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>88.5</td>
<td>10.2</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>64.4</td>
<td>9.8</td>
</tr>
</tbody>
</table>
7.4.2 Radon gas in air within a room having 0.5 circulations/h

Figure 7.5 Five different concrete samples and their calculated radon gas rates within a room as a function of Relative Humidity (RH).

7.4.3 Radon gas in air within a room having 0.5 circulations/h

Figure 7.6. Five different concrete samples and their calculated radon gas rates as a function of time (weeks) after initiation of casting the concrete.
7.5 Phase III – 0.45 and 0.65 w/c ratio using one crushed aggregate

7.5.1 General

In the third phase, two concrete samples were assessed with the same aggregate but different w/c ratios. The investigations also measured the difference in weight of the two concrete samples as curing progressed.

7.5.2 Radon exhalation rate and mass (kg) as a function of Relative Humidity (RH)

In Table 7.5 the measured and calculated exhalation rates are presented in conjunction with a link to the phase II study in regard to the aggregates used. The calculations of the radon gas within the room are shown with the ventilation rate of 0.5 circ./hour (Figure 7.7). The loss in mass (kg) is shown in Figure 7.8.

Table 7.5 Measured and calculated exhalation rates of each concrete sample as well as the w/c ratio.

<table>
<thead>
<tr>
<th>Id-nummer</th>
<th>Bedrock</th>
<th>Link to phase II</th>
<th>RH</th>
<th>Radongas exhalation rate (Bq/m²h)</th>
<th>w/c ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sample 1</td>
<td>Granitoid/ gneiss</td>
<td>Sample 3</td>
<td>97.00</td>
<td>39.40</td>
<td>0.65</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>94.10</td>
<td>30.10</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>91.50</td>
<td>34.10</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>85.50</td>
<td>24.00</td>
<td></td>
</tr>
<tr>
<td>Sample 2</td>
<td></td>
<td>Sample 3</td>
<td>92.50</td>
<td>37.40</td>
<td>0.45</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>87.60</td>
<td>37.70</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>83.20</td>
<td>26.40</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>76.00</td>
<td>21.70</td>
<td></td>
</tr>
</tbody>
</table>

Figure 7.7 Radon exhalation rate (Bq/m²h) as a function of the Relative Humidity (%). Two different w/c ratios of the concrete castings were measured.
Figure 7.8 Mass in kg of the concrete samples as a function of the Relative Humidity (%). Measurements were initiated 1 day after casting the concrete samples.

7.6 Phase IV – Combined results from phase I and II – 0.5 circ/h

A comparison of the calculated data was commenced for four of the samples used in the different phases. The calculated data from Phase I and II were compared since the same aggregates have been used for the concrete specimens, where only their w/c ratios are different. The combined data are presented in Figure 7.9.

Figure 7.9. Calculated radon gas in air as a function of Relative Humidity for a circulation of 0.5 circ./h of eight concrete samples where four were assessed in phase I and four in phase II.
7.7 Link between $^{222}$Rn (radon gas) and $^{226}$Ra ($^{238}$U) gamma radiation.

As could be seen from Figures 7.5 and 7.9 the radon gas levels within a room are fairly constant at RH below 80-75 %. Figure 7.10 presents a link between a given exhalation rate, recalculated to radon gas contribution within a room and the gamma radiation of $^{226}$Ra ($^{238}$U).

For a proper estimation of the exhalation rate of the concrete specimens, the following approach was adopted. A mean value of the last three readings, for sample 1 to 4 presented in Table 7.4 served as a basis for each individual concrete sample’s “fixed” exhalation rate.

In the Figure (7.10), two rates of ventilation within the room in relation to the $^{226}$Ra content of the concrete sample are exemplified.

![Graph showing the relationship between Radon gas (Bq/m³) and $^{226}$Ra (Bq/kg) with linear equations $y = 1.9743x$ and $y = 1.1963x$.](image)

**Figure 7.10.** Data from four of the concrete samples, where the calculated radon gas content of the specified room, given a specific ventilation rate (circ./h), is shown as a function of the $^{226}$Ra content of the samples (values from laboratory analysis - gamma spectrometer – STUK).

7.8 Uncertainties in the calculations of radon gas

Calculations according to the ISO-standard ISO-11665-7 include the margin for errors (uncertainties). The estimated standard deviation $s$ has been calculated for two sets of repeatable measurements. In percent a variation of 4.2-6.4 % was established. However, due to the fairly limited number of repetitions (see Section 7.9) a cautious approach should be adopted. Thus, to be conservative, in Figure 7.11 a presentation of uncertainty bars including a 10 % deviation from the measured value is accounted for. The operational uncertainties have been considered negligible.

Figure 7.11 presents some of the results of the phase II study.
Figure 7.11 Calculated values of the radon gas in air for a specified room including the standard deviation $s$ (10%) as a function of time (weeks). Samples 1, 4, 5 are plotted – part of the phase II study (w/c ratio of 0.65).

### 7.8.1 Repeatability of measurements

The repeatability of the measurements was checked three times (Table 7.6). In the first phase, two measurements were performed. The second time, three measurements of the same concrete sample was executed consecutively. And the third time, five measurements were performed in the same manner. In all cases an RH below 60% of the concrete samples had been established. Between the measurements, the radon gas container was circulated with air from the outside for at least several hours in order to ensure low initial values $C_0$ of radon gas.

The standard deviation ($s$) as well as uncertainty ($u$) with a coverage factor of $k=1$ (~68% confidence level) were calculated according to Eq. 7.2 and 7.3 (Bell, 2001):

$$s = \sqrt{\frac{\sum_{i=1}^{n} (x_i - \bar{x})^2}{n-1}} \quad (7.2)$$

$$u = \frac{s}{\sqrt{n}} \quad (7.3)$$
Table 7.6. Calculated exhalation rates according to ISO 1165-7 including the standard deviations (s) and the uncertainty (u) with a coverage factor of \( k = 1 \).

<table>
<thead>
<tr>
<th>Set I</th>
<th>Exhalation rate (Bq/m²h)</th>
<th>Mean (exhalation rate)</th>
<th>Standard deviation (s) (±%)</th>
<th>Uncertainty (u %)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sample I</td>
<td>36.9</td>
<td>34.7</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>32.5</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Sample II</td>
<td>9.7</td>
<td>9.47</td>
<td>0.4</td>
<td>4.2</td>
</tr>
<tr>
<td></td>
<td>9</td>
<td></td>
<td></td>
<td>2.4</td>
</tr>
<tr>
<td>Sample III</td>
<td>44.4</td>
<td>46.78</td>
<td>3.0</td>
<td>6.4</td>
</tr>
<tr>
<td></td>
<td>47.3</td>
<td></td>
<td></td>
<td>2.9</td>
</tr>
<tr>
<td></td>
<td>50.0</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>43.0</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>49.2</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

7.8.2 Reproducibility

The reproducibility was checked at SP – the Unit of Sustainable Built Environment by using analogous equipment as CBI and with an identical alpha-particle counter (ATMOS 33). In Table 7.7 the results are presented.

Table 7.7. Calculated exhalation rates (Bq/m²h) after measuring the same concrete specimen using two different set ups, but with an identical radon monitor.

<table>
<thead>
<tr>
<th>Set I</th>
<th>Exhalation rate (Bq/m²h)</th>
<th>Mean (exhalation rate)</th>
<th>Standard deviation (s)</th>
<th>(±%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sample I</td>
<td>36.9*</td>
<td>37.1</td>
<td>0.3</td>
<td>0.8</td>
</tr>
<tr>
<td></td>
<td>37.3**</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

*Measured at CBI, **Measured at SP – Sustainable Built Environment
8 Discussions
This Thesis deals with ionizing radiation in concrete buildings and has primarily focused on:

I. Clarification of quantities, regulations, units and deriving values for specific conversion factors.
II. Measurement techniques of ionizing radiation of the natural radionuclides using a hand-held spectrometer, laboratory gamma spectrometry and geochemical analysis using ICP-MS/EOS.
III. Description of a European model (EC, 1999) and its use to calculate the ionizing radiation of aggregates and concretes within a predefined room by using the I-index.
IV. A quick non-destructive methodology for determination of gamma radiation and radon gas exhalation rate using a concrete portion with a specified recipe.
V. Investigation of the relation between relative humidity and radon gas, using a fix porosity value and different w/c ratios.
VI. Calculation of the radon gas contribution of concrete in a specified room model, using different ventilation rates.

8.1 Gamma radiation

8.1.1 Sources of errors
Gamma energies in relation to construction products (building materials) could reasonably well be defined with a gamma spectrometric hand-held equipment. But knowledge on the effects that may influence the specific energies is vital to correctly evaluate the result. Stahls et al. (2014) demonstrate this phenomenon by comparing the mass of the object in relation to the efficiency of the instrument. The surface area and more importantly the volume of the whole object are essential in relation to portable equipment used of the construction product. Especially due to its calibration and the conditions, that encompasses the tested object.

Most likely this is one of the causes, as described above, why the portable hand held equipment, used by CBI, shows an activity (Bq/kg) slightly less than the analytical procedures at higher values (Chapter 6, Figure 6.1). The concrete specimen areas may be slightly too small to register the full energy in relation to the area of the calibration site and the given geometries. To highlight this, on four occasions, materials were sampled in the field, and sent for gamma spectrometric evaluation (STUK). Measurements were made within two meters of the sampling area on flat surfaces. The results sent for gamma spectrometric analysis indicated minor difference for two of four samples (Chapter 6, Table 6.3). In one case a very good correlation with the laboratory gamma spectrometry even at high values was demonstrated. Hence, this implies, that most likely, in order to achieve a consistent reading of the materials examined, the large concrete specimens (1.5 × 1.5 m) would have needed to be even larger due to the large area from which the field equipment absorbs gamma rays, when calibration occurs.

Secondly, and in part in contrast to the above stated, there may also be an excess in energy of the samples analyzed due to very fine fractions analyzed. This could in part be visualized in Paper III where in Table 2, the specific activity (Bq/kg) of ⁴⁰K, ²²⁶Ra and ²³²Th each analyzed particle size fraction are presented. It can be noted, that for each case, the finer fractions show an increase in the specific activity (Bq/kg) of the above mentioned radionuclides compared to the coarser fractions (meaning, larger portion of fines in the material generates slightly higher values). The procedure to crush in order to homogenize has been used for every sample. However, as demonstrated, this may also give rise to slightly higher values at the laboratories than would be the normal case if measured on a more coarse fraction, as is the case measuring the concrete specimens.

The thickness of the concrete specimens is yet another variable, which to some extent also contributes to lower values registered by the field spectrometer of CBI (Chapter 6, Figure 6.7). This has also been demonstrated by Hoffman (2014) presenting a dose model for the calculation of effective dose within a room. However, the effect on a flat surface should be limited to a maximum of a few percent

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compared to a thickness of 300-350 mm, which is considered as a maximum of a normal concrete wall thickness, where the gamma emitting nuclides are equally hindered by self-absorption as to contribute to the gamma emitting energies of the construction material (TC/WG3/TG32 technical draft).

Furthermore, it has to be noted that all concrete slabs were measured in a state of RH > 90, while for the analytical laboratories the samples, were dried and measured, which in turn may yield a minor increase in radiation (Ishimori et al, 2013) of the nuclides.

Finally, the secular equilibrium conditions are always assumed using the portable hand-held equipment. As demonstrated in Paper I, plots of the specific activity of $^{40}$K, $^{226}$Ra and $^{232}$Th of the same concrete materials using the different analyzing techniques are shown, which indicates that a secular equilibrium condition of the radionuclides may well not have existed for at least one large concrete specimen measured.

Using geochemical analysis the sample size (the small portion), that is analyzed is sometimes a hindrance as for proper evaluation. This is also emphasized in Paper I, due to errors of results from initial measurements sent for analysis.

All in all, the fairly good agreement between the geochemical and gamma spectrometric analysis justifies both methods as viable. Secondly, and in favour of the laboratory analysis is that the influences of the background and cosmic radiation are controlled. However, for values which are not in close range of a critical reference values, the portable hand-held spectrometer serves its purpose as an excellent instrument and should be justified to use in many assessments, even for measurements on smaller areas as demonstrated within this Thesis.

8.1.2 Correlation of results in relation to theoretical calculations

Investigations on how to calculate the effective dose within specified conditions of a room, are still related to theoretical calculations as demonstrated by Markkanen (1995) as well as Hoffman (2014) and Nuccitelli et al. (2015). The final outcome of the effective dose, at the end, also includes dependencies of several different construction products, put on top (sandwich construction including concrete, gypsum boards, tiles, wall paper), which make calculations difficult. In the light of this, the Thesis has focused on a simplified case where only concrete as a construction product is discussed. Erlandsson & Isksson (2006) and Almgren et al. (2008) have shown the ability of measuring a correct personal dose within dwellings, however, without knowing the initial conditions of the materials used. Meaning, this assessment is the final state of measuring the personal dose ($H_{p(10)}$), which could be justified as the effective dose (ICRP 2007) of the construction product placed within the building. Thus, in order to justify impact of the building materials assessed within this Thesis a final step is lacking – verification of the theoretical calculations and empirical analysis of the building materials gamma emitting levels, measured as personal dose, when the building material is part of a room.

However, some relations could be established by use of the current document RP 112 and its suggested I-index. The I-index has also been assessed in relation to some international indices (Hex, Hin, Raeq), more specifically presented in Paper II. By and large, most indices are comparable and give a similar indication of a building materials risk to produce naturally ionizing radiation to inhabitants. In Paper III there are assessed theoretical calculations of a building material, e.g. aggregates constituting part of the concrete with an I-index of ~1 that would yield an effective dose of ~0.65 mSv per year, using a standard concrete recipe as presented within the text (Chapter 6, Table 6.5) and a recently presented dose assessment by Hoffman (2014). But, this is, as described in the former section, the result of the sole product; concrete. The strong r-values (Garcia, 2010) show the dependence of the I-index of aggregates in relation to the calculations made from the analyzed specific activities of the radionuclides (Bq/kg) of the concretes using the dose model by Hoffman to calculate the final dose.

Theoretically calculated values of the I-index of the cast concretes (Chapter 6, Table 6.6) was also validated. The calculations were made by analyzing each constituent`s content of natural radioactivity
(40K, 226Ra, 232Th). The theoretical calculations were thereafter compared to the analyzed results of the concrete mixes. In all cases the difference between the theoretically calculated I-index and the I-index calculated from the results of the concrete mix was less than 15 %, except for one sample (sample four). Sample four displayed a difference of more than 50 % and this could only be explained by improper sampling. Sample seven (~14 % difference) and in part samples 10 and 11 (~9 % difference) could in part be related to variations in the aggregates (not homogenously distributed) within the concrete or in part aggregate variations when sampled from the woven bags.

But overall, out of the eleven samples assessed, a median value of 2.4 % in difference between theoretical calculations and analyzed results of the concrete mixes does imply an overall good agreement between theory and analyses. The author’s opinion is that this has to be viewed as acceptable in the light of uncertainties for the concrete mixing process and uncertainties of each constituent analyzed using the applied accredited test-method.

8.1.3 Comparative data of the CBI methodology

This Thesis has also shown that a very good proportionality could be established between smaller concrete samples and larger concrete specimens with a given thickness, using a portable hand-held spectrometer and a lead-shielded box for measurements. The strong R²-values indicate a strong trustworthiness in the correlated values (Garcia, 2010). Thus, the utilization of the non-destructive methodology put forward as to calculate the I-index is seen as reliable. Using the same concrete recipe for other aggregates, this proposed methodology can be utilized to quite accurately calculate the final ambient dose rate, H*(10) or I-index of the concrete specimens.

8.1.4 Consequences of the results and explanations

The Thesis has partly tried to show that by use of different constituents as part of the final building material (concrete) the building material could be tuned with respect to the final specific activity (Bq/kg) often expressed as I-index. The importance of decreasing the radioactive content of the building materials is in relation to reference values put forward by the European Commission (EC 1999, 2014). This process has been demonstrated through theoretical calculations and the assessment of empirical data of activity concentrations measured between aggregates and when the very same aggregates constitute part in the concrete (Paper III).

A consequence of the result presented, a commencement to optimize many building materials in regard to natural ionizing radiation of its constituents, may be initiated. The prime objective shall be as to reduce the natural radioisotopes as low as possible within the final building material. The Thesis has in part tried to aid in this area as to achieve this objective. Meaning, ways to reduce the gamma radiation of the building materials could as presented (Chapter 6) be achieved by shifting constituents or at least replacing parts of them.

8.2 Radon gas

8.2.1 Sources of errors

Sources of errors are related to the calculation procedure and directly linked to the measurement technique used were an initial “zero” value have to be established. The “zero”-value is in Eq. 5.2 (Section 5.4) defined as C₀. The value is calculated as a mean out of the last six readings, but easily varies with a factor of two depending on the radon exhalation rates in the atmosphere (air taken from the outside). At lower to medium exhalation rates of the concrete samples, this “zero-value” may influence the final exhalation rate strongly and has to be reflected upon, when analyzing the object or a larger set of data.
The instrumental technique using a pulsating ionization chamber developed by Baltzer et al. (1992) has a certainty of 10% (s.d). The repeatability measurements of two concrete samples also imply that this is a valid relation indicating a variation of 4-6% between measurements.

8.2.2 Comparative data

Initial calculations using a w/c ratio of 0.45 implied for some concrete mixes an overall increase of radon gas exhalation rate in the interval RH 85-65% as the humidity decreased. However, at RH < 65% the trend was shifted to a more stabilized value. For concrete mixes showing lower exhalation rates the trend was indistinct. This latter observation is in line with measurements performed by de Jong et al. (1996) where no clear trend could be observed. De Jong et al. (1996), however, measured concrete with very low exhalation rates.

The concept of using a “twin”-concrete sample, in the phase I study, as to measure the RH was time consuming and not very sophisticated. At the end, the results are doubtful, likely due to this practice, since RH is not measured on the actual analyzed concrete sample, even though, the concrete samples “parent” and “daughter” were treated identically.

However, the second attempt using a higher water-cement ratio (0.65), where plastic tubes for measurement of RH were inserted within 24-72 hours after molding, show a distinct trend. According to de Jong et al. (1996) an increased w/c ratio would yield an increased hydration rate and consequently an increase in capillary pores. Seemingly, this may apply. The rate of exhalation or the radon gas content of air (Bq/m³) is, in the phase II-study, clearly decreasing, for each concrete sample investigated. Specially, this could be seen in the interval of 98-80% in RH. Thereafter each trend becomes more constant as RH decreases. Convincingly, the radon exhalation rates of the concrete portions are influenced by relative humidity, at least at high RH-values. The latter results are in line with investigation of van der Graaf (2003), where also a downtrend of the exhalation rate was modeled for RH < 80-90%. However, there is no evidence, within this study of a low exhalation rates above 90% in RH as modeled by van der Graaf et al. (2003), rather the opposite has been shown.

The current results of the study also support earlier investigations made by Petterson et al. (1982), where moisture, relative humidity and exhalation rate were evaluated. Although only a few (two samples) concrete specimens were investigated, a decrease of 10% in exhalation rate was concluded, starting with a relative humidity of ~90% and dropping to ~50%.

The third phase was launched to validate the results of the second phase. Seemingly, the outcome is in line with the results reported for w/c ratios of 0.65. But also, the concrete sample with a w/c ratio of 0.45 shows a similar trend, as the one reported in phase II. The measured radon exhalation rate of this concrete sample also decreases in the interval of 95-85% in RH.

Even though the uncertainties of the measurements are rather high (10%), the trend seen in Figure 7.9 and highlighted in Section 7.11 (Chapter 7) could not be excused by measurement uncertainties.

As a result of the latest data, the implications are that there may be doubtfulness to the first phase of results (w/c ratio 0.45). But, as could be seen in Chapter 7, Figure 7.3 or 7.9 most of the measurements did not commence (due to the process of sawing concrete samples out of the large concrete specimens) before or after the concrete portions had an RH of ~85-80%. Thus the most vital changes, in exhalation rate of the concrete samples may have occurred prior to the commencements of the measurements.
8.2.3 Consequences of the results and explanations
The calculations of the content of radon gas (Bq/m\(^3\)) using the exhalation rate from the surface of concrete ISO 11665-7 (2012) in relation to calculations of ventilation rates and radon gas production within a specified room (Åkerblom and Clavensjö, 2007) have shown that there are strong influences of the exhalation rate at the beginning of the hydration process. This needs to be considered before measurements are performed within a building.
9 Conclusions

9.1 Gamma radiation

Below there is a summary of conclusions drawn from the study of gamma radiation in relation to the building materials investigated.

- This Thesis has demonstrated some different analyzing techniques that can be used to calculate the gamma energies of a building material as well as the final construction product’s gamma energy using I-index and a dose model.

- The portable hand-held spectrometer is a useful tool for measurements but knowledge of its calibration, the volume that is measured and geometry surrounding the equipment are vital factors affecting the calculated results. The I-index could be well estimated for values less than 1 for the investigated cast concrete specimens.

- By use of different analytical techniques very strong correlation coefficients (>0.9 in r-value) have been demonstrated between an aggregate and when this aggregate is part of a concrete recipe.

- The study has also demonstrated the fragility of the decay chain, using radioisotopes where secular equilibrium has to be assumed. This is valid for both the hand-held spectrometer and the laboratory gamma spectrometry by STUK.

- The construction product used for prefabricated purposes, with a cement content of 350 kg/m³ and a water-cement ratio of 0.45 and a content of aggregates constituting ~80 % in mass, will yield a loss of approximately 16-19 % in I-index compared to its measured aggregates. The effective dose using calculations by Hoffman has been assessed. A difference of approximately 35 % between the measured aggregate and the final effective dose to the human body has been established. This is usable for an I-index of 1 for the aggregates, assuming an approximate recipe as described in Table 4.6 and a concrete thickness of 200 mm as well as density of ~2350 kg/m³ of the concrete.

- The thickness has a rather large implication between 150 and 350 mm (~20 %) within a room, but has only a limited influence between 150 and 200 mm, measured on a plane surface. An increase of 2-5 % in absorbed dose, H*(10), was demonstrated by CBI on a plane surface.

- The influence of density within this project and the densities of the aggregates used (2260-2950 kg/m³) has a very limited influence on the effective dose in the dose model by Hoffman (2014).

- This Thesis has also demonstrated how to calculate the final activity concentration (Bq/kg) within a concrete specimen using different building materials with known gamma energies of $^{40}$K, $^{226}$Ra and $^{232}$Th.

- A proposed methodology was presented as a non-destructive method to calculate the activity of a large concrete slab making use of a small concrete portions and correlation data established during the project for 100, 150 and 200 mm thick concretes.

- The hand-held spectrometer has been presented explaining some features of its internal calculation process in relation to protection and monitoring quantities and units used for regulatory purposes.
9.2 Radon gas

The investigations in relation to radon gas revealed some important features such as:

- The w/c ratio seemingly plays a significant role in the characteristics of the radon exhalation rate development as RH decreases, especially at RH above 80-85 %.

- Concrete cubes with a w/c ratio of 0.65 showed a steady decrease in radon exhalation rate until 80-70 % in RH was reached and thereafter stabilized at lower RH ratios.

- Dubious results were achieved for the first setup using w/c ratio of 0.45.

- Due to the above facts, measurements of radon gas within a building should not be performed during the first year.

- Calculation of the radon gas content within a room is largely affected by the ventilation rate. Using 0.5 circulations/h yield results, where six out of eight different concrete mixes have values below the stipulated reference level of 200 Bq/m$^3$.

- A solid correlation was established between the $^{226}$Ra content and the $^{222}$Rn gas in air within a room, using different ventilation rates, calculating a mean value of exhalation rates between 80 and 60 in RH.

- Repeatability measurements and reproducibility measurements of the radon exhalation rate yielded stronger correlations than expected.
10 Future research

Future research is multifold and the author believes that a better connection between theoretical values and practical values of the finalized dwelling has in part to be improved. There is as of today no clear practical link between construction products put on the market for declaration of performance and their final contribution to the natural radiation when measured in a dwelling, due to the fact that the product is one of many within the building. Hence a stronger verification process would be optimal where confirmation of the theoretical values calculated by the I-index or dose model could be assessed in a finalized building. One way to approach this would be to construct a room, with the specified conditions as described by EC (2014) in order to verify the final results in effective dose, presented as personal dose (Hp)10 in relation to theoretical values of effective dose calculated within this project.

Secondly, the laboratory gamma spectrometry technique as used, within this project, and as recommended by the EC, still has disadvantages using secondary equilibrium of the primordial radionuclides. Other techniques, such as neutron activation of the natural radioisotopes (40K, 226Ra, 232Th) should perhaps be investigated further to achieve a more precise result of some building materials.

The exhalation rate of radon gas as a function of relative humidity in concrete implies that much effort could be done to better control the rate of exhalation and possibly lower it. Chauhan & Kumar (2013) and Taylor-Lange et al. (2012) have showed that input of different additives, such as fly ash or silica to some degree lowers the exhalation rate. This is a necessity to further explore, due to the environmental regulations put forward by clients today, where radon gas levels in dwellings of < 50 Bq/m³ is awarded in public procurements. Hence, this encourages further research of how to reduce the radon gas exhalation rates by use of new additives as radon closures, whilst being a part of the concrete.

Finally, Al-Jarallah et al (2001) address the wide spread of results measuring radon gas using active and passive techniques and making comparative analysis. The results are discouraging. This would not be acceptable if threshold values < 50 Bq/m³ in radon gas are to be assessed and proven. Consequently, also measurement techniques, on how to swiftly and accurately evaluate radon content need to be recognized, evaluated and improved.
11 References


Appended papers