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Gamma exposure from building materials – a dose model with expanded gamma lines from Naturally Occurring Radionuclides applicable in non-standard rooms.

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Keywords

Euratom Basic Safety Standards; Dose Models; Naturally Occurring Radionuclides; Naturally Occurring Radioactive Materials; Radioactivity; Building materials; Concrete; Inorganic polymers; Alkali-activated materials; External gamma exposure

Abstract

Building materials are a significant source of gamma rays exposure due to the presence of naturally occurring radionuclides. In order to protect the public from harmful radiation, the European Basic Safety Standards (Council directive 2013/59/Euratom) introduced a one-size-fits-all building(s) (materials) Activity Concentration Index (ACI) based on a limited set of gamma lines. The ACI is considered “as a conservative screening tool for identifying materials that may cause the reference level (i.e. 1 mSv/y) laid down in Article 75(1) to be exceeded”. Regarding calculation of dose, many factors such as density and thickness of the building material, as well as factors relating to the type of building, and the gamma emission data need to be taking into account to ensure accurate radiation protection. In this study the implementation of an expanded set of 1845 gamma lines, related to the decay series of ²³⁸U, ²³⁵U and ²³²Th as well as to ⁴⁰K, into the calculation method of Markkanen [1], is discussed. The expanded calculation method is called the Expanded Gamma Dose Assessment (EGDA) model. The total gamma emission intensity increased from 2.12 to 2.41 and from 2.41 to 3.04 for respectively the ²³⁸U and ²³²Th decay series. In case of ⁴⁰K a decrease from 0.107 to 0.1055 is observed. The ²³⁵U decay series is added, having a gamma emission intensity of 3.1. In a standard concrete room, the absorbed dose rates in air (D_A) per unit of activity concentration of 0.849, 0.256, 1.08, 0.0767 nGy/h per Bq/kg are observed. The use of weighted average gamma lines increased the D_A with 6.5 % and 1 % for respectively the ²³⁸U and ²³²Th decay series. A decrease of 4.5 % is observed in the D_A of ²³⁵U decay series when using the weighted average gamma lines in comparison to its non-averaged variant. The sensitivity of the EGDA model for density, wall thickness, presence of windows and doors and room size is investigated. Finally, a comparison of the index and dose calculations relevant for the dose

47 assessment within the European legislative framework applicable towards building materials
48 is performed. In cases where the ACI and density and thickness corrected dose calculation of
49 Nuccetelli et al. [2] cannot provide guidance, the EGDA allows performing more accurate dose
50 assessment calculations leading to effective doses which can be several 100 $\mu\text{Sv/y}$ lower.

51

52 **1. Introduction**

53

54 Building materials are a significant source of indoor gamma dose [3]. The importance to
55 address the exposure originating from building materials is underlined in article 75 of the
56 Euratom basic safety standards (EU-BSS) (Council directive 2013/59/Euratom) which comes
57 into force in February 2018 [4]. This article states that "*The reference level applying to indoor
58 external exposure to gamma radiation emitted by building materials, in addition to outdoor
59 external exposure, shall be 1 mSv per year*". This European legislation was developed to
60 establish basic standards, applicable in EU member states, for the protection against exposure
61 of ionising radiation for workers and the general public. In a broader context this legislation
62 supports several launched initiatives of the European commission for turning waste into a
63 resource and promoting re-use and recycling with focus on the building industry in the
64 framework of the Europe 2020 strategy [5–7]. In this context the EU-BSS aims towards a safe
65 use of by-products, originating from NORM (Naturally Occurring Radioactive Material)-
66 processing industries, like metallurgical slags, fly and bottom ash, phosphogypsum and red
67 mud. These residues are used or investigated to use in cement-based matrixes as
68 supplementary cementitious materials (SCM) on a large scale [8–12]. In addition more and
69 more research is conducted to use these residues in more CO₂-friendly cement alternatives,
70 like inorganic polymers (IPs) [8–10]. This fits with the aim to reduce the usage of primary
71 resources. It is expected that future building materials used for dwellings will shift more and
72 more towards these secondary raw materials that can potentially be rich in naturally occurring
73 radionuclides (NORs): therefore the impact on the external gamma exposure of the use of
74 these secondary raw materials needs to be assessed [10,13,14].

75

76 In order to assess the impact on external gamma exposure of building materials, different
77 calculation methods, based on Monte Carlo simulations, integration and simple index and
78 dose formulas, have been developed in the past [1,15–26]. Different dose assessment
79 calculations have been developed based on gamma ray attenuation and build-up factors
80 [1,16,17,22,27]. These calculations allow specifying the physical parameters of the room and
81 the material it is constructed out, in a straightforward way. The density and wall thickness are
82 identified as the most critical parameters. Modifying these parameters, for the evaluation of
83 non-standard rooms, can generate dose rate differences up to 40 % compared to a standard
84 concrete room [27]. Seeking for a standardized approach, the EU-BSS proposes a screening
85 index, named Activity Concentration Index (ACI) [2]. This index was originally developed by
86 Markkanen [1] and is described in the technical guide Radiation Protection (RP)-112 [28]. The
87 ACI is based on a number of assumptions that are not all necessarily valid. The ACI assumes a
88 concrete room (400 cm x 500 cm x 280 cm) with a density of 2350 kg/m³ and thickness of 20
89 cm for all surfaces (walls, floor and ceiling). In the last years, in order to get a reliable screening
90 tool, that will allow for a realistic discrimination of building materials, a new density and
91 thickness corrected index I(p_d) was developed by Nuccetelli et al. [2]. The available dose
92 assessment models focus on the standard composition of concrete, however the increased
93 usage of residues, which have an *a priori* chemical compositions differing from conventional

94 raw materials (like OPC and gravel), can result in structures with very different compositions.
95 Some models consequently apply a correction factor to compensate for the different
96 composition [29]. In addition, disequilibrium in the ^{238}U and ^{232}Th decay series chain can be
97 present for residues from NORM-processing industries. Information regarding disequilibrium
98 can be valuable for gaining insight into environmental or industrial processes. However, when
99 dealing with the dose assessments of building materials one should assess how meaningful
100 the consideration of disequilibrium is. Up to now, to the authors' knowledge, in none of the
101 existing dose calculations, disequilibrium situations are taken into account. In contrast RP-122
102 [30] suggests using the highest activity concentration of a radionuclide present in a certain
103 decay series to specify the activity concentration of that whole decay series. In none of the
104 existing tools the presence of ^{235}U and its decay products is considered.

105
106 The above mentioned calculation methods have in common that they only use a fraction of
107 the gamma emission lines known today. In practice, this means that often dose models use a
108 specific set of major gamma lines or that the set of several major gamma lines is reduced to
109 one or several averaged gamma lines with the gamma intensity as weighing factor. Whereas
110 the gamma emission intensity of this averaged gamma line is the sum of the individual gamma
111 emission intensities. This technique is performed to provide simplicity. However progress has
112 been made in the characterization of the gamma emissions of radionuclides. The Laboratoire
113 National Henri Becquerel has built an online database providing continuously updated
114 information on the gamma emission lines of a wide range of radionuclides that allows going
115 beyond this simplified approach [31]. Implementation of this database into a dose calculation
116 method allows a more accurate safety assessment to evaluate if construction products can be
117 used from a radiation protection point of view [2]. Both sample parameters, like density and
118 composition, as well as room parameters like thickness of the walls, ceiling and floor, number
119 of walls present, the sample composition of each wall etc. impact the final received dose
120 [15,27]. An adaptable dose assessment calculation allows taking these parameters into
121 account.

122
123 Using an flexible dose or index calculation, in contrast to a screening index, for the evaluation
124 of building materials fits better with the 1 mSv dose requirement of article 75 of the EU-BSS
125 [2], in particular when dealing with non-standard room and building material parameters. In
126 addition the implementation of non-standard room and building material parameters deals
127 with the requirement of annex VII of the EU-BSS, that states "*The calculation of dose needs to
128 take into account other factors such as density, thickness of the material as well as factors
129 relating to the type of building and the intended use of the material (bulk or superficial)*". The
130 current study implements improvements, based on scientific data available in literature, into
131 the existing and validated Markkanen room model. A sensitivity analysis of the different
132 parameters impacting the calculated absorbed dose rate in air is performed. For the different
133 improvements implemented in the dosimetric evaluation the impact and practicality for
134 industrial implementation is discussed.

135 136 **2. Materials & Methods**

137 138 **2.1 Materials**

139

140 For the evaluation of the dose model the composition of concrete, defined by NIST [32], is
 141 used, except when mentioned differently.

142

143 2.2 Model

144

145 2.2.1 Model description

146

147 To assess the absorbed dose rate in air (D_A), the room model of Markkanen [1] (see Equation
 148 1) is used.

149

150

$$151 \quad D_A = 5.77 \times 10^{-7} \frac{AC\rho}{4\pi} \sum \gamma_i \left(\frac{\mu_{en}}{\rho} \right)_i E_i \int B_i \frac{e^{-\mu_i s}}{l^2} dV \quad (1)$$

152

153

154 With D_A the absorbed dose rate in air in Gy/h, AC the activity concentration of a radionuclide
 155 incorporated in the material of concern in Bq/kg, ρ the density of the material in kg/m³, γ_i the
 156 gamma intensity of gamma line i , $(\mu_{en}/\rho)_i$ the energy absorption coefficient in air for gamma
 157 energy E_i in cm²/g, E_i the photon energy in MeV, μ_i the linear attenuation coefficient of the
 158 material for gamma energy E_i in cm⁻¹, B_i the dose build up factor (see Equation 2) calculated
 159 via the Berger's formula, l the distance between the point of detection (x_p, y_p, z_p) and the point
 160 of integration in cm (see Equation 4) and s the fraction of l within the top layer in cm (see
 161 Equation 3). The total exposure rate is the sum of the exposure rates calculated from ceiling,
 162 floor and each wall. The $(\mu_{en}/\rho)_i$ is a polynomial best fit achieved from the data reported by
 163 Martin [33] using the data of Hubbell and Seltzer [34].

164

$$165 \quad B_i = 1 + C(E_i)\mu_i s e^{D(E_i)\mu_i s} \quad (2)$$

166

167 In literature different C and D parameters are proposed by different authors. In the model
 168 described here, the values of C and D proposed by Pelliccioni [35] are used. These are
 169 calculated for the energy spectrum via logarithmic and exponential best-fit function
 170 respectively by using the concrete parameters described by Pelliccioni [35] at 7 mean free
 171 paths (mfp).

172

$$173 \quad s = \left| \frac{z}{z_p - z} \right| l \quad (3)$$

174

$$175 \quad l = \sqrt{(x_p - x)^2 + (y_p - y)^2 + (z_p - z)^2} \quad (4)$$

176

177 In order to convert the D_A to effective dose a conversion factor of 0.7 Sv/Gy is used [28]. This
 178 conversion factor is used for all gamma emitters and originates from the UNSCEAR 2000 report
 179 [3].—This conversion factor is used in the dose calculations considered in this article and is
 180 consequently used for comparison reasons. Nevertheless, nuclide specific conversion factors
 181 have been suggested by Krstic and Nikezic [36].

182

183 The model assumes a homogeneous sample composition and a homogeneous distribution of
184 the radionuclides throughout the composed materials. In addition a standard room is used as
185 a reference throughout the paper. The standard room size was described by Koblinger [15] as
186 measuring 400 cm x 500 cm x 280 cm. Here a standard thickness of walls, floor and ceiling of
187 20 cm is assumed. Neither doors nor windows are present and the point of detection (x_p , y_p ,
188 z_p) is set at the middle of the room. Whereas Koblinger suggested a density of 2320 kg/m³, RP-
189 112 suggests a density of 2350 kg/m³ [15,28]. The value of 2350 kg/m³ is used here as a
190 standard.

191 No background correction is assumed when calculating the D_A .

192

193 All calculations are performed by a combination of Microsoft® excel and R® [37]. The input
194 parameters are submitted in Microsoft® excel whereas the further treatment of the input data
195 is performed by Microsoft® excel and R®.

196

197 2.2.2. Selection of the number of gamma lines

198

199 In order to check the impact of the number of gamma lines, a comparison of the absorbed
200 dose rate in air is made between different dose assessment models for a standard room. The
201 Markkanen [1], Mustonen [22], ISS room model [23] and the model developed in this study,
202 further called Expanded Gamma Dose Assessment (EGDA) model, are compared. Different
203 versions of the EGDA model are evaluated depending on the number of gamma lines used for
204 the dose assessment. 'EGDA>1%', 'EGDA>0.1%', 'EGDA>0%' take into account all gamma lines
205 which have a gamma emission intensity (including the branching factor) above respectively 1
206 %, 0.1 % and 0 % when considering gamma emission lines from the ²³⁸U, ²³²Th and ²³⁵U decay
207 series and ⁴⁰K. In addition two variants of 'EGDA>0.1%' are discussed. In one variant the
208 emission gamma lines of ²³⁸U and ²³²Th (except for the 2614 keV gamma emission line since
209 this emission line represents over 40 % of the dose rate of the ²³²Th decay series) of
210 'EGDA>0.1%' are converted to one weighted average gamma emission line. This variant is
211 indicated in Table 1 by the suffix "averaged". In the second variant the emission gamma lines
212 which have a gamma emission intensity lower than 0.1 % are converted to one weighted
213 average gamma line for ²³⁸U, ²³²Th and ²³⁵U. This variant is indicated in Table 1 as "EGDA+".
214 Details on each model are provided in Table 1. Since not all the details necessary for the
215 calculations were present in the original paper of Markkanen [1] and Mustonen [22], updated
216 values were used (details in Table 1). This is indicated by a suffix "updated". In addition, a
217 second variant of the ISS room model, which makes use of the Berger parameters described
218 by Pelliccioni [35] instead of the Berger parameters of Markkanen [1], is discussed. This variant
219 is indicated with the infix "Pelliccioni" whereas the original ISS room model is indicated with
220 the infix "original". For readability, abbreviations of the dose model names are provided in
221 Table 1 and Table 5.

222

223 Table 1: Overview of the different dose calculation models and their parameters used to
 224 evaluate the absorbed dose rate in air.

Model	Markkanen original	Markkanen updated	Mustonen updated	ISS original room model	ISS Pelliccioni room model	EGDA>1%	EGDA>0.1%	EGDA>0.1% averaged	EGDA+	EGDA>0%
Concrete composition	Markkanen 1995 [1]	Ordinary Portland concrete (NIST)	Ordinary Portland concrete (NIST)	Ordinary Portland concrete (NIST)	Ordinary Portland concrete (NIST)	Ordinary Portland concrete (NIST)	Ordinary Portland concrete (NIST)	Ordinary Portland concrete (NIST)	Ordinary Portland concrete (NIST)	Ordinary Portland concrete (NIST)
Energy absorption coefficient in air	Markkanen 1995 [1]	Best fit from Martin 2006 [33]	Best fit from Martin 2006 [33]	Hubbell 1982 [38]	Hubbell 1982 [38]	Best fit from Martin 2006 [33]	Best fit from Martin 2006 [33]	Best fit from Martin 2006 [33]	Best fit from Martin 2006 [33]	Best fit from Martin 2006 [33]
Density (kg/m³)	2350	2350	2350	2350	2350	2350	2350	2350	2350	2350
Linear attenuation coefficient	From Markkanen 1995 [1]	XCOM [39]: ordinary Portland concrete (NIST)	XCOM [39]: ordinary Portland concrete (NIST)	Hubbell 1982 [38]	Hubbell 1982 [38]	XCOM [39]: ordinary Portland concrete (NIST) or IP	XCOM [39]: ordinary Portland concrete (NIST) or IP	XCOM [39]: ordinary Portland concrete (NIST) or IP	XCOM [39]: ordinary Portland concrete (NIST) or IP	XCOM [39]: ordinary Portland concrete (NIST) or IP
Gamma emission energy and intensity	Markkanen 1995 [1]	Mustonen 1984 [22]	Mustonen 1984 [22]	NuDat website [40]	NuDat website [40]	DDEP website [31]	DDEP website [31]	DDEP website [31]	DDEP website [31]	DDEP website [31]
Berger Parameters	Markkanen 1995 [1]	Best fit of Pelliccioni 1989 [35]	Best fit of Pelliccioni 1989 [35]	Best fit of Markkanen 1995 [1]	Best fit of Pelliccioni 1989 [35]	Best fit of Pelliccioni 1989 [35]	Best fit of Pelliccioni 1989 [35]	Best fit of Pelliccioni 1989 [35]	Best fit of Pelliccioni 1989 [35]	Best fit of Pelliccioni 1989 [35]
Number of gamma lines ²³⁸U	1	1	24	19*	19*	82	87	1*	87 + 1**	761
Gamma emission intensity ²³⁸U***	2.12	2.12	2.12	2.41	2.41	2.19	2.36	2.36	2.41	2.41
Number of gamma lines ²³²Th	2	2	20	14*	14*	36	110	2*	110 + 1**	349

Gamma emission intensity $^{232}\text{Th}^{***}$	2.41	2.41	2.41	2.63	2.63	2.76	2.98	2.98	3.04	3.04
Number of gamma lines ^{40}K	1	1	1	1	1	1	1	1	1	1
Gamma emission intensity ^{40}K	0.107	0.107	0.107	0.107	0.107	0.1055	0.1055	0.1055	0.1055	0.1055
Number of gamma lines ^{235}U	-	-	-	-	-	47	128	1*	128 + 1**	734
Gamma emission intensity $^{235}\text{U}^{***}$	-	-	-	-	-	2.78	3.04	3.04	3.1	3.1
Model abbreviation	Mark _{orig}	Mark _{upd}	Must _{upd}	ISS _{orig}	ISS _{Pelli}	-	-	-	-	-

* 87, 109 and 128 gamma emission lines are converted to 1 for respectively ^{238}U , ^{232}Th and ^{235}U

** 674, 239 and 606 gamma emission lines are converted to 1 for respectively ^{238}U , ^{232}Th and ^{235}U

*** The gamma emission intensity is the sum of the individual gamma-ray emission energies.

225

226 2.2.3. Role of the build-up factor

227

228 The impact of the build-up factor (B) was evaluated for a standard room by using different
 229 sets of Berger parameters C and D to calculate the D_A per unit of activity concentration. The
 230 Berger parameters as described by Markkanen and by Pelliccioni were compared [1,35]. In
 231 addition the case without Berger parameters (C=D=0) is evaluated, meaning the role of build-
 232 up factor is neglected. The latter case is indicated by the suffix "B = 1" in Table 4.

233

234 2.2.4. Role of the presence disequilibria in the ^{232}Th , ^{238}U and ^{235}U decay series

235

236 For model 'EGDA>0.1%' the contribution of long living radionuclides and their progeny to the
 237 total absorbed dose rate in air per unit of activity concentration for the decay series of ^{238}U
 238 and ^{232}Th is evaluated. The ^{238}U decay chain is divided into 3 subchains : i.e. ^{238}U -part (^{238}U to
 239 ^{230}Th), ^{226}Ra -part (^{226}Ra to ^{214}Po) and ^{210}Pb -part (^{210}Pb to ^{210}Po). Similar, the ^{232}Th decay chain
 240 is divided into ^{232}Th -part (only ^{232}Th), ^{228}Ra -part (^{228}Ra to ^{228}Ac) and ^{228}Th -part (^{228}Th to ^{208}Tl).
 241 The absorbed dose rate in air of ^{235}U is evaluated in the framework of the ratio of AC of
 242 $^{238}\text{U}/^{235}\text{U}$ i.e. 21.6 as expected value for non- diluted/enriched samples. No disequilibrium is

243 considered in case of ^{235}U decay series as (dis)equilibrium in this decay series is often not
244 reported.

245

246 2.2.5 Impact of sample specific composition

247

248 The impact of the sample composition on the dose rate is compared by simulating a room
249 constructed out of Fayalite Slag based Inorganic Polymers (FSIPs). FSIPs have different
250 chemical, physical and structural properties than concrete. The characteristics of FSIPs are
251 described by Kriskova et al. [41], Onisei et al. [42] and Iacobescu et al. [43]. The sample
252 composition differs from concrete consequently leading to the usage of different linear
253 attenuation coefficients. The attenuation coefficients are calculated for each gamma emission
254 energy via the XCOM program [44]. The sample specific coherent mass attenuation coefficient
255 of XCOM is therefore converted to the sample specific linear attenuation coefficient.

256

257 **2.3. Sensitivity Analysis**

258

259 A sensitivity analysis of the parameters impacting the absorbed dose rate in air is performed.
260 The studied parameters are density, wall thickness, presence of windows and doors and room
261 size. All parameters are compared to the standard parameters of a standard concrete room
262 as defined in section 2.2.1.

263

264 2.3.1. Density

265

266 The impact of the wall density on the D_A is tested for a standard room with density varying
267 stepwise (step size of 100 kg/m^3) between 1000 kg/m^3 and 3500 kg/m^3 , corresponding to the
268 density of hollow bricks up to the density of high density concrete.

269

270 2.3.2. Wall Thickness

271

272 In a standard concrete room the wall thickness is assumed to be 20 cm. However depending
273 on the usage thinner or thicker walls are required. The impact of the wall thickness on the D_A
274 in the standard room is tested with wall thickness varying stepwise (step size of 5 cm) between
275 5 cm and 80 cm while keeping floor and ceiling thickness constant at 20 cm.

276

277 2.3.3. Room Size

278

279 The impact of the room size on the D_A is tested for a concrete room. A square room is
280 simulated with length of the wall varying stepwise (step size of 100 cm) between 100 cm and
281 1000 cm for a room height of 280 cm and between 100 cm and 1183.2 cm for a room height
282 of 200 cm.

283

284 2.3.4 Presence of windows and doors

285

286 The EU-BSS assumes a standard room without the presence of windows and doors. This is a
287 strict approach but not realistic. The impact of the presence of windows or doors of different
288 surfaces is tested. Tests are conducted for surfaces of 1 m^2 , 2 m^2 and 4 m^2 positioned in the

289 middle or the corner of a wall or ceiling. The imaginary dose rate originating of the specific
 290 window/door surface is subtracted from the dose rate of the wall without any window/door.

291

292 **2.4 Comparison of index and dose assessment tools**

293

294 A comparison is made of the most used index and dose calculations relevant for the dose
 295 assessment within the European legislative framework applicable towards building materials.
 296 More details regarding these different index and dose calculations are shown in Table 2 or can
 297 be found in the respective references.

298

299 The index values calculated via ACI and the density and thickness corrected index (I(pd)) are
 300 compared using the AC of different types of residues and cement shown in Table 3 [1,2,4]. The
 301 obtained dose of the Markkanen original, density and thickness corrected (D(pd)) and
 302 EGDA>0% dose calculations are compared using the same AC [1,2,30]. In addition to the
 303 standard density of 2350 kg/m³ and standard thickness of 20 cm, six different scenarios are
 304 tested with varying density and thickness (Table 4). In the comparisons, it is assumed that the
 305 residues are solely used to construct a building material, this because recent studies [45,46]
 306 indicate the applicability of building materials without the use of any additives like cement,
 307 sand, gravel, etc. The AC values originate from Nuccetelli et al. [10]. In all cases the exposure
 308 time is 7000 h.

309

310 Table 2: Overview of the parameters of the index and dose calculations used in the European
 311 legislative framework applicable towards building materials.

	Index calculation		Dose calculation		
	ACI	I(pd)	Markkanen original	D(pd)	EGDA>0%
Geometry	Floor, ceiling, 4 walls	Floor, ceiling, 4 walls	Floor, ceiling, 4 walls	Floor, ceiling, 4 walls	Floor, ceiling, 4 walls
Size geometry (cm³)	400 x 500 x 280*	400 x 500 x 280	400 x 500 x 280	400 x 500 x 280	(Flexible) Here 400 x 500 x 280
Wall thickness (cm)	20	Flexible	20	Flexible	Flexible
Density (kg/m³)	2350**	Flexible	2350**	Flexible	Flexible
Background correction	70 nGy/h	50 nGy/h	0.348 mSv	0.245 mSv	0.245 mSv

Composition	Concrete	Concrete	Concrete	Concrete	(Flexible) Here Concrete
Reference(s)	EC 2014; RP112; Markkanen 1995	Nucetelli et al. 2015	EC 2014; RP112; Markkanen 1995	Nucetelli et al. 2015	

* In Markkanen 1995 size is 12 x 7 x 2.8 m³ with thickness of 0.2 m

** In Markkanen 1995 density is 2320 kg/m³

312

313 Table 3: Activity concentrations (Bq/kg) of ²²⁶Ra, ²³²Th and ⁴⁰K present in different residues

314 and cement.

Material type	²²⁶Ra (Bq/kg)	²³²Th (Bq/kg)	⁴⁰K (Bq/kg)	Reference
Furnace slags *	147	42	258	Nucetelli et al. 2015
Bottom ash and fly ash *	207	80	546	Nucetelli et al. 2015
Phosphogypsum *	381	22	71	Nucetelli et al. 2015
Bauxite residue *	337	480	205	Nucetelli et al. 2015
Cement *	42	32	214	Nucetelli et al. 2015

315

* Average values of database from Nucetelli et al. 2015 [10]

316

317 Table 4: Description of 6 different scenarios which are described by a specific set of density

318 and thickness. The scenarios are used for the comparison of the models of Table 2.

319

Scenario number	1	2	3	4	5	6
Thickness (cm)	10	10	18	25	40	40
Density (kg/m³)	1400	3000	3000	1400	1400	3000

320

321

322 **3. Results and discussion**

323

324 In section 3.1 the different absorbed dose rates in air per unit of activity concentration for
325 ²³⁸U, ²³²Th, ⁴⁰K and ²³⁵U obtained by different dose assessment models are compared. This
326 section discusses the impact of working with averaged gamma emission lines as well as the
327 impact of the build-up factor and the radiological equilibria.

328

329 Based on this comparison, the most practical EGDA model with the highest gamma emission
330 intensity is selected and in section 3.2 a sensitivity analysis of this model is performed by
331 changing wall thickness and density, room size and the presence of windows and doors.
332 Throughout section 3.1 and 3.2 the impact of the sample composition is quantified.

333

334 Section 3.3 deals with the application of the model focussing on the dosimetric evaluation,
335 the impact and the practicality for industrial implementation. Consequently a comparison is

336 performed of the most used index and dose calculations relevant for the dose assessment
 337 within the European legislative framework applicable towards building materials.

338

339 **3.1. Model**

340

341 **3.1.1. Impact of the number of gamma lines**

342

343 Table 5 shows the D_A per unit of activity concentration for ^{238}U , ^{232}Th , ^{40}K and ^{235}U of the
 344 different models described in Table 1. The different models assume a concrete standard room
 345 unless indicated else by suffix FSIP.

346

347 Table 5: Overview of the absorbed dose rate in air per unit of activity concentration (nGy/h
 348 per Bq/kg) for ^{238}U , ^{232}Th , ^{40}K and ^{235}U calculated by different dose assessment calculation
 349 models described in Table 1.

350

	^{238}U	^{232}Th	^{40}K	^{235}U
Model abbreviation	Dose rate in air (D_A) (nGy/h per Bq/kg)	Dose rate in air (D_A) (nGy/h per Bq/kg)	Dose rate in air (D_A) (nGy/h per Bq/kg)	Dose rate in air (D_A) (nGy/h per Bq/kg)
Mark _{orig}	0.908	1.06	0.0767	-
Mark _{orig B=1}	0.3845	0.5	0.0408	-
Mark _{upd}	0.893	1.02	0.0778	-
Mark _{upd B=1}	0.383	0.501	0.0407	-
Must _{upd}	0.84	0.999	0.0778	-
Must _{upd B=1}	0.0405	0.51	0.0407	-
ISS _{orig}	0.894	1.138	0.0767	-
ISS _{Pelli}	0.869	1.109	0.0767	-
EGDA>1%	0.76	0.967	0.0767	0.228
EGDA>0.1%	0.826	1.06	0.0767	0.25
EGDA>0.1% _{B=1}	0.395	0.535	0.0401	0.0819
EGDA>0.1% FSIP	0.838	1.07	0.0784	0.234
EGDA>0.1% _{aver}	0.88	1.07	0.0767	0.239
EGDA>0.1% _{aver B=1}	0.368	0.51	0.0401	0.0725
EGDA+	0.85	1.08	0.0767	0.255
EGDA>0%	0.849	1.08	0.0767	0.256

351 Suffix 'orig' (original): Data of the original paper are used as shown in Table 2.

352 Suffix 'upd' (updated): Updated data, as shown in Table 2, are used with the original calculation method.

353 Suffix 'B=1' (build-up factor = 1): The Berger parameters are set to zero. This means the role of the build-up
 354 factor is negligible.

355 Suffix 'aver' (averaged): Several gamma lines are reduced to a single weighted average gamma emission line.

356 Suffix 'Pelli' (Pelliccioni): the Berger parameters as described by Pelliccioni 1989 are used.

357 FSIP: The chemical composition of the room components is set to the FSIP chemical composition.

358

359 Comparing the D_A between $Mark_{orig}$ and $Mark_{upd}$, an increase of 1.7 % and 3.8 % is observed
360 for respectively ^{238}U and ^{232}Th , in favour of the $Mark_{orig}$ model. In case of ^{40}K a decrease of 1.4
361 % is observed in favour of the $Mark_{orig}$ model. This deviation in D_A is due to the usage of
362 different Berger parameters and a different concrete composition in the two models (Table
363 1).

364
365 The 24 emission gamma lines of ^{238}U and the 19 gamma emission lines (2614 keV-line is
366 excluded) of ^{232}Th of the Mustonen model are converted to a single weighted average gamma
367 emission line for ^{238}U and ^{232}Th in the Markkanen model.

368 Comparing $Mark_{upd}$ with $Must_{upd}$ a 6 % and 2 % increase in D_A is observed for respectively ^{238}U
369 and ^{232}Th . This increase is solely due to usage of averaged gamma lines in the Markkanen
370 model. In case of ^{235}U a decrease in the D_A of 4.6 % (4.9 %) is observed for the 'averaged EGDA'
371 variant. The differences are solely due to the usage of energy specific attenuation coefficients
372 and energy specific C and D Berger parameters as the total gamma intensity stays equal.

373
374 When comparing the EGDA models with $Mark_{upd}$, $Must_{upd}$ and the ISS room models one can
375 see that the number of gamma lines used is much higher (Table 1). When more gamma lines
376 are included in the EGDA model the gamma emission intensity also increases for ^{238}U , ^{232}Th
377 and ^{235}U , leading to higher D_A when comparing EGDA>1%, EGDA>0.1% and EGDA>0%.
378 However the gamma emission intensity of the ISS room model is smaller than the gamma
379 emission intensity of EGDA>0% for ^{238}U and ^{232}Th (Table 1), still the D_A of the ISS room model
380 is higher than the D_A of EGDA>0% (Table 5). The usage of a set of averaged gamma-lines in the
381 ISS room models tends to increase the D_A , as discussed above. In addition the usage of other
382 B in the ISS_{orig} (Table 1) also impacts the D_A , this is discussed in section 3.1.2.

383
384 The EGDA>0% model uses all the gamma lines available originating from ^{238}U , ^{232}Th , ^{235}U and
385 ^{40}K . In total 1845 gamma lines are used in the calculation by model EGDA>0% whereas in
386 model EGDA>0.1% 326 gamma lines are used. The gamma emission intensity of EGDA>0.1%
387 is 2.1 %, 2.0 % and 1.9 % lower than EGDA>0% for respectively ^{238}U , ^{232}Th and ^{235}U .
388 Nevertheless, when using a higher number of gamma lines also the calculation time increases.
389 In order to limit the calculation but still consider the maximum gamma emission intensity, the
390 extra gamma lines of EGDA>0% in comparison to EGDA>0.1% are converted to 3 weighted
391 average gamma lines; one line for ^{238}U , ^{232}Th and ^{235}U . This approach is incorporated in the
392 EGDA+ model (Table 1). The difference in D_A between EGDA+ and EGDA>0% is limited to plus
393 0.001 nGy/h per Bq/kg for ^{238}U and minus 0.001 nGy/h per Bq/kg for ^{235}U . In case of ^{232}Th no
394 difference was observed.

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396 3.1.2. Impact of the Build-up factor

397

398 Table 5 shows the D_A for several models. Comparing the D_A of the "B=1" variants with the non-
399 unity originals, a significant decreases in the D_A is present. For example in the case of
400 EGDA>0.1% the "B=1" variant has an D_A which is approximately 52 %, 50 %, 48 % and 67 %
401 lower for respectively ^{238}U , ^{232}Th , ^{40}K and ^{235}U . The presence of the B is consequently
402 important when calculating the D_A . The ISS_{Pelli} model differs only from the ISS_{orig} model by the
403 usage of the data of Pelliccioni instead of the data of Markkanen to calculate the B. Comparing
404 both models, the D_A per unit of activity concentration of the ISS_{Pelli} model is 2.8 % and 2.6 %
405 lower for respectively ^{238}U and ^{232}Th in case of a standard concrete room.

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3.1.3. Impact of disequilibrium in the ^{232}Th , ^{238}U and ^{235}U decay series

Table 6: Absorbed dose rate in air (D_A) per unit of activity concentration (nGy/h per Bq/kg) of the long-living radionuclides and their progeny of the ^{238}U and ^{232}Th decay series in case of the EGDA>0.1% model.

Concrete standard room					
^{238}U Decay series			^{232}Th Decay series		
	D_A (nGy/h per Bq/kg)	% Contribution		D_A (nGy/h per Bq/kg)	% Contribution
^{238}U Part	0.0077	0.931	^{232}Th Part	0.000041	0.004
^{226}Ra Part	0.82	99.002	^{228}Ra Part	0.42	39.583
^{210}Pb Part	0.00055	0.067	^{228}Th Part	0.64	60.413

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Considering the decay series of ^{238}U : the ^{238}U -part, ^{226}Ra -part and ^{210}Pb -part of the decay chain represent respectively approximately 0.93 %, 99 % and 0.067 % of the total external absorbed gamma dose rate in air per unit of activity concentration of the whole ^{238}U decay series, in the case of a standard concrete room. The lifespan of a building material will not allow reestablishing the equilibrium between the ^{238}U -part and ^{226}Ra -part. Looking solely at the lifespan aspect, it would be meaningful to treat both parts of the decay chain separately. However, this is not always feasible since one must be able to measure ^{238}U , ^{234}Th or ^{234}Pa . Using in this case the AC of ^{226}Ra for the whole decay series will only introduce a small bias since ^{238}U part and ^{210}Pb contribute less than 1 % to the total D_A of the ^{238}U decay series. On the other hand using the AC of ^{238}U for ^{226}Ra and its decay products would have a large impact as ^{226}Ra -part represents 99 % of the D_A of the ^{238}U decay series. The suggestion of RP-122 to use the highest AC present in the decay series would overestimate the gamma dose rate when the AC of ^{238}U or ^{210}Pb is larger than the AC of ^{226}Ra . Due to the small contribution of ^{210}Pb -part to the gamma dose (i.e. 0.067%), the activity concentration of ^{226}Ra is used for the ^{210}Pb -part of the decay series in this study. The half-life of ^{222}Rn allows radon exhalation from the building material which decreases the external absorbed gamma dose rate in air. De Jong and Van Dijck (2008) [18] showed that the external absorbed gamma dose rate in air decreased on average with 9 % and 5 % for respectively gypsum and concrete used in the Netherlands. In addition the EU-BSS [4] treats the radon exposure (from soil and building materials) separately from the gamma exposure linked to building materials. For this reason all the EGDA models do not consider radon and is therefore stricter in terms of gamma ray exposure.

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Considering the decay series of ^{232}Th : the ^{232}Th -part, ^{228}Ra -part and ^{228}Th -part of the decay chain represent respectively approximately 0.004 %, 39.6 % and 60.4 % (Table 6) of the total external absorbed gamma dose rate in air per unit of activity concentration of the whole ^{232}Th decay series in the case of a standard concrete room. Disequilibria in the ^{232}Th decay chain are complex and insights in the production process of NORM-residues can provide useful information. In the case of complete Th-separation, the equilibrium will install within a timeframe of 40 years in the Th-bearing residue. Whereas in the Ra-bearing residue the activity will fade away. The lifetime of building materials can be considered to cover this timespan. Being strict, it is best not to consider disequilibrium and consider the highest activity concentration that is possible and use for the complete (so 100 %) D_A calculation of the ^{232}Th

446 decay series. An adequate determination of the activity concentration is recommended to
447 assess whether or not disequilibria are present. In addition it is assumed in this study that no
448 ^{220}Rn exhalation from the building material takes place as the half-life of ^{220}Rn is relatively
449 short (55.8 sec).

450

451 To the authors knowledge in none of current dose assessments tools available, the decay
452 series of ^{235}U is considered. However taking into account all the gamma emission intensities
453 above 0.1 % the absorbed dose rate in air is 0.250 nGy/h per Bq/kg for a standard concrete
454 room (Table 5). This is above the D_A of ^{40}K on a Bq/kg level. However framing this ^{235}U D_A in a
455 broader context, when the natural abundance of U is respected the AC of ^{235}U is 0.0463 times
456 the AC of ^{238}U . So in reality the contribution of the D_A of ^{235}U is of limited consequence, except
457 when high activity concentrations of ^{238}U are present. When no ^{235}U is measured, the authors
458 recommend using 0.0463 times the AC of ^{238}U to implement the dose originating from ^{235}U .
459 Within the ^{235}U decay series, disequilibrium situations can also be present but these are not
460 considered here.

461

462 3.1.4. Impact of sample specific composition

463

464 The impact of the sample composition is studied by comparing EGDA>0.1% and EGDA>0.1%
465 FSIP. An increase in the D_A of 1.4 %, 0.9 % and 2.1 % is observed for ^{238}U , ^{232}Th and ^{40}K when
466 FSIP is used instead of concrete. On the contrary, in case of ^{235}U , a decrease in the D_A of 6.8 %
467 is observed. It has to be noted that here solely the linear attenuation coefficients are changed.
468 A change of sample composition implies also changing the energy and mfp-dependent B, due
469 to the interdependency between the composition, the energy and the mfp. However, the
470 study of this aspect is outside the scope of this paper.

471

472 3.1.5. Selection of EGDA>0% model

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474 The EGDA>0% model uses the highest gamma emission intensity and makes use of all the
475 nuclear data on an individual base. Consequently this approach is the more accurate one and
476 is selected for the performance of a sensitivity analysis in section 3.2. ~~The use of 3 weighted
477 gamma emission lines in case of ^{238}U , ^{232}Th and ^{235}U , corresponding to respectively 2.1 %, 2.0
478 % and 1.9 % of the total gamma emission intensity, allows performing faster calculations in
479 comparison to EGDA>0% model.~~ The C and D Berger parameters described by Pelliccioni
480 (1989) [35] are used for the calculations.

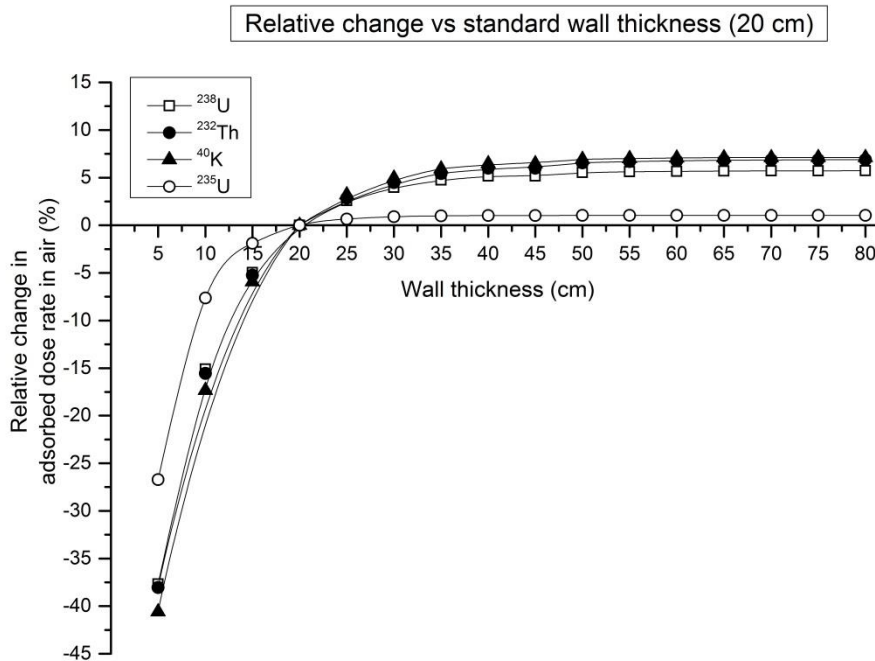
481 The presence of gamma emission by ^{235}U is considered and disequilibrium situations can be
482 considered when necessary.

483

484 3.2. Sensitivity Analysis of EGDA>0% Model

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486 3.2.1. Impact of the wall thickness calculated by the EGDA>0% model.



487
 488 Figure 1: Relative change in the absorbed dose rate in air (D_A) for a standard concrete room
 489 with varying thickness (5-80 cm) vs a standard concrete room with wall thickness of 20 cm for
 490 ^{238}U , ^{232}Th , ^{40}K and ^{235}U . Relative change: $(D_{A\text{thickness}_X} - D_{A\text{thickness}_{20\text{cm}}}) / (D_{A\text{thickness}_{20\text{cm}}} \times$
 491 100)

492 Figure 1 shows the relative change (%) in D_A air for different thicknesses relative to the wall
 493 thickness of 20 cm for ^{238}U , ^{232}Th , ^{40}K and ^{235}U in a standard concrete room. It is observed that
 494 the relative decrease in D_A occurs rapidly with decreasing wall thickness. In case of a wall
 495 thickness of 5 cm a relative decrease of 27.4 %, 27.6 %, 28.9 % and 21.1 % is observed for
 496 respectively ^{238}U , ^{232}Th , ^{40}K and ^{235}U . In case of a wall thickness of 80cm a relative increase of
 497 6.1 %, 7.4 %, 7.7 % and 1.1 % is observed for respectively ^{238}U , ^{232}Th , ^{40}K and ^{235}U . However, a
 498 plateau in the increase of the D_A is observed. The percentage increase of the D_A between 20
 499 cm and 25 cm thickness is below 1 % for ^{235}U whereas for the other radionuclides this is
 500 approximately 3 %. From a thickness of 40 cm, the increase in the D_A is below 1 % per increase
 501 in 5 cm thickness for all the radionuclides. According to Risica et al. (2001) [27] this plateau
 502 originates from self-absorption effects.

503

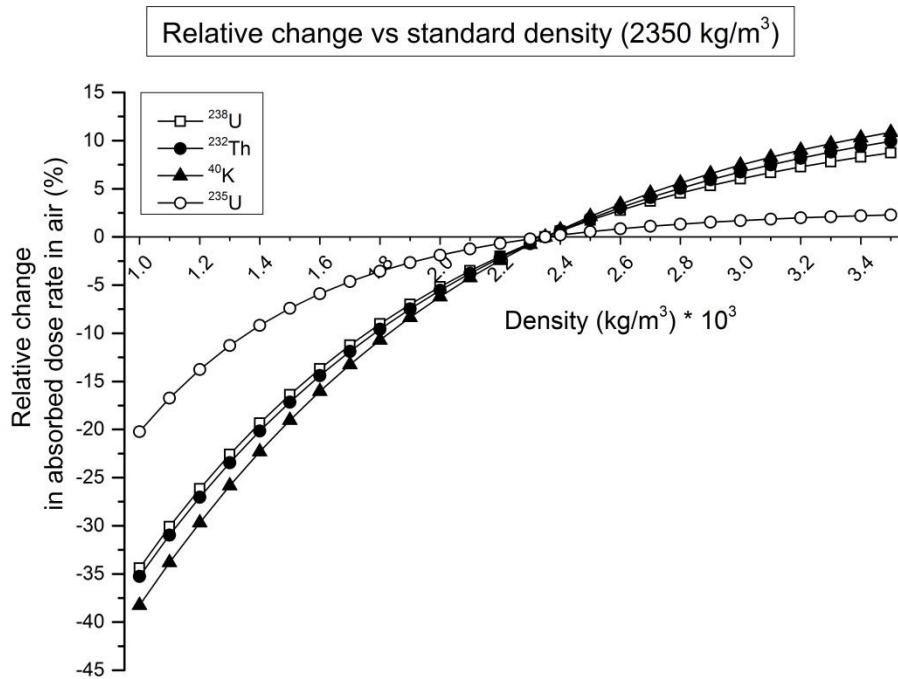
504 As the floor thickness is not varied the contribution of the walls to the D_A will increase with
 505 the thickness. The contribution of the smaller wall (400 cm) will increase with approximately
 506 5 % relative to the larger wall (500 cm) when increasing the wall thickness from 5 cm to 80
 507 cm.

508

509 3.2.2. Impact of the density calculated by EGDA0% model

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514 Figure 2: Relative difference of the absorbed dose rate in air (D_A) for a standard concrete room
515 with varying density (1000-3500 kg/m^3) vs a standard concrete room with density of 2350
516 kg/m^3 for ^{238}U , ^{232}Th , ^{40}K and ^{235}U . Relative change: $(D_{A\text{density}_X} - D_{A\text{density}_{2350 \text{ kg/m}^3}}) /$
517 $(D_{A\text{density}_{2350 \text{ kg/m}^3}} \times 100)$

518

519 Figure 2 shows the difference in D_A for different densities relative to the standard density of
520 2350 kg/m^3 for ^{238}U , ^{232}Th , ^{40}K and ^{235}U in a standard concrete room with thickness of 20 cm.
521 At densities lower than 2350 kg/m^3 a relative decrease in D_A is observed whereas a relative
522 increase is observed at densities higher than 2350 kg/m^3 . In case of a density of 1000 kg/m^3
523 a relative decrease of 34 %, 35 %, 38 % and 20 % is observed for respectively ^{238}U , ^{232}Th , ^{40}K and
524 ^{235}U . In case of a density of 3500 kg/m^3 cm a relative increase of 9 %, 10 %, 11 % and 2 % is
525 observed for respectively ^{238}U , ^{232}Th , ^{40}K and ^{235}U . With increasing densities the total number
526 of radionuclides present in the material will increase leading to higher D_A . With decreasing
527 densities to contrary is true.

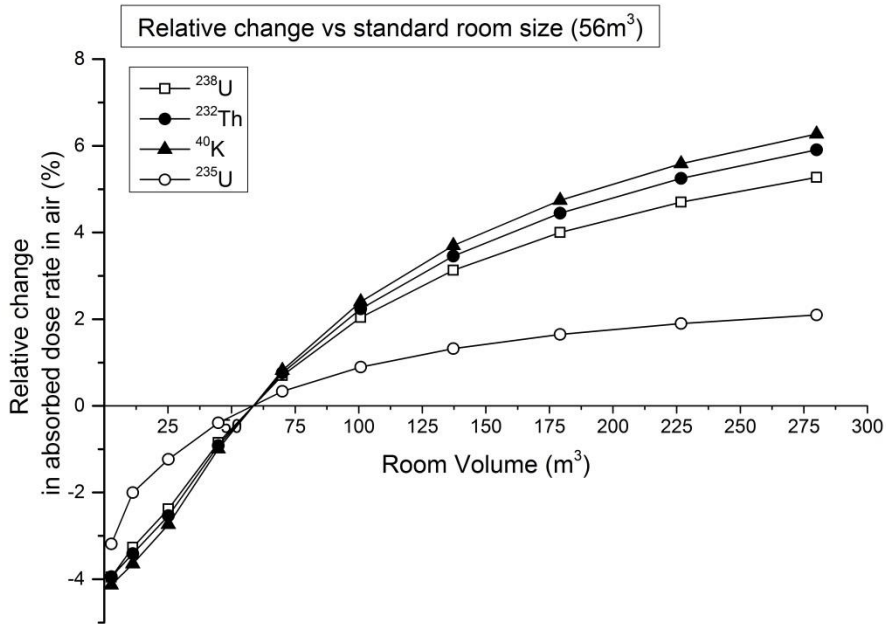
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529 With increasing densities the relative contribution of the floor and ceiling to total dose rate
530 decreases with approximately 1 % whereas the dose rate of the walls increases slightly. This
531 effect is observed for the different radionuclides.

532

533 3.2.3. Impact of the room size calculated by the EGDA>0% model

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537 Figure 3: Relative difference of the absorbed dose rate in air for a standard concrete room
538 with varying room size (2.8 - 280 m³) vs a standard concrete room with room size of 56 m³ for
539 ²³⁸U, ²³²Th, ⁴⁰K and ²³⁵U.

540

541 Figure 3 shows the difference in D_A for different room sizes relative to the standard room size
542 (200 x 250 x 280 cm³) for ²³⁸U, ²³²Th, ⁴⁰K and ²³⁵U in a standard concrete room. It is observed
543 that the relative decrease in D_A occurs with decreasing room size. In case of a room size of 2.8
544 m³ a relative decrease of 4 %, 4 %, 4 % and 3 % is observed for respectively ²³⁸U, ²³²Th, ⁴⁰K and
545 ²³⁵U. In case of a room size of 280 m³ a relative increase of 5 %, 6 %, 6 % and 2 % is observed
546 for respectively ²³⁸U, ²³²Th, ⁴⁰K and ²³⁵U. With increasing room size to person standing in the
547 room is surrounded by more material. Consequently the total number of radionuclides
548 present in the room will also increase, leading to higher D_A. With decreasing room size the
549 contrary is true.

550 Figure 3 also shows that the influence of the room sizes affects the radionuclides differently.

551

552 Next to changing the room surface the impact of the room height is studied. At small room
553 volumes (below approximately 15 m³), the D_A of ²³²Th is lower in case of a height of 200 cm
554 than in case of a height of 280 cm. For a room area of 1 m² a difference of approximately 2.3
555 %, 2.5 %, 2.7 % and 1.2 % difference for respectively ²³⁸U, ²³²Th, ⁴⁰K and ²³⁵U is observed.
556 However at room size larger than 15 m³ the impact of height on the D_A is reverted. At a room
557 volume of 280 m³ an increase in the D_A of 1.7 %, 2.0 %, 2.1 % and 0.5 % for respectively ²³⁸U,
558 ²³²Th, ⁴⁰K and ²³⁵U is observed in favour of the room height of 200 cm .

559

560 3.2.4 The impact of the presence of windows and doors by the EGDA>0% model

561

562 Table 7: % Deviation in dose rate for different window surfaces located in the middle or the
 563 corner of Wall 1 (400cmx280cm), Wall 2 (500x280cm) and the ceiling (400 cm x 500cm) in
 564 comparison to respectively Wall 1, Wall 2 and the ceiling without the presence of windows.
 565

	Window 100 cm x 100 cm		Window 100 cm x 200 cm		Window 200 cm x 200 cm	
	Middle	Corner	Middle	Corner	Middle	Corner
% Deviation in absorbed dose rate in air (D_A) originating from wall 1 (400 cm x 280 cm) with a window in comparison wall 1 without a window						
^{238}U	-11.8	-7.3	-22.5	-21.1	-43.2	-36.6
^{232}Th	-11.8	-7.3	-22.5	-21.1	-43.1	-36.6
^{40}K	-11.7	-7.4	-22.4	-21.0	-43.0	-36.6
^{235}U	-12.1	-7.2	-23	-21.4	-43.9	-36.7
% Deviation in absorbed dose rate in air (D_A) originating from wall 2(500 cm x 280 cm) with a window in comparison to wall 2 without a window						
^{238}U	-11.7	-4.5	-21.9	-19.9	-41.1	-26.4
^{232}Th	-11.6	-4.5	-21.8	-19.9	-41.0	-26.4
^{40}K	-11.5	-4.6	-21.7	-19.8	-40.7	-26.4
^{235}U	-12.2	-4.3	-22.6	-20.4	-42.2	-26.1
% Deviation in absorbed dose rate in air (D_A) originating from a ceiling (500 cm x 400 cm) with a window in comparison to a ceiling without a window						
^{238}U	-14.6	-2.2	-22.8	-6.4	-40.8	-17.3
^{232}Th	-14.4	-2.3	-22.5	-6.4	-40.4	-17.2
^{40}K	-13.9	-2.2	-21.7	-6.3	-39.2	-16.9
^{235}U	-15.7	-2.1	-23.9	-6.1	-42.5	-16.9

566 Table 7 shows the percentage of deviation in the D_A of the different room components in
 567 comparison to the standard concrete room. With increasing size of the window or door
 568 surface the D_A of the component decreases. For example, in wall one the D_A decreases with
 569 approximately 12 % in case of a window of 100 cm x 100 cm whereas this decrease is
 570 approximately 37 % for a window of 200 cm x 200 cm. In both cases the windows are
 571 positioned in the middle of wall. Nevertheless the position of the surface in the component
 572 plays an important role. In wall 2 the D_A decreases for approximately 41% when the window
 573 is positioned in the middle of the wall. When the same window is positioned in the corner, the
 574 D_A decreases with 26 % in comparison to a standard concrete room. In addition it must be
 575 noted that in the case of a standard concrete room wall 1, wall 2 and the floor/ceiling
 576 contribute for approximately 9.5 %, 14.5 % and 26 % respectively to the total D_A of the room.
 577 The final influence on the D_A due to the presence of a window in the ceiling will be larger than
 578 for a window in wall 1.
 579

580
 581 **3.3 Comparison of index and dose calculations**
 582

583 Table 8: Overview of the index-values and effective dose (mSv/y) of the index and dose
 584 calculations used in the European legislative framework for different building materials
 585 consisting of residues or cement.
 586

	Index							
Model	ACI	I(pd)						
Thickness (cm)	20	10	10	18	20	25	40	40
Density (kg/m ³)	2350	1400	3000	3000	2350	1400	1400	3000
Furnace slags	0.788	0.384	0.628	0.811	0.770	0.678	0.822	1.006
Bottom ash and fly ash	1.269	0.609	0.997	1.290	1.225	1.077	1.307	1.602
Phosphogypsum	1.405	0.719	1.171	1.510	1.434	1.264	1.529	1.864
Bauxite residue	3.592	1.657	2.710	3.509	3.330	2.928	3.554	4.355
Cement	0.385	0.180	0.295	0.382	0.363	0.319	0.387	0.476
Scenario number		1	2	3		4	5	6
	Dose (mSv/y)							
Model	Mark _{orig}	D(pd)						
Thickness (cm)	20	10	10	18	20	25	40	40
Density (kg/m ³)	2350	1400	3000	3000	2350	1400	1400	3000
Furnace slags	0.726	0.238	0.549	0.745	0.704	0.606	0.755	0.916
Bottom ash and fly ash	1.293	0.521	1.017	1.329	1.264	1.108	1.346	1.604
Phosphogypsum	1.592	0.659	1.237	1.595	1.521	1.342	1.614	1.905
Bauxite residue	3.825	1.841	3.190	4.043	3.865	3.437	4.087	4.796
Cement	0.206	-0.019	0.128	0.222	0.202	0.155	0.227	0.304
Scenario number		1	2	3		4	5	6
	Dose (mSv/y)							
Model	EGDA>0%							
Thickness (cm)		10	10	18	20	25	40	40
Density (kg/m ³)		1400	3000	3000	2350	1400	1400	3000
Furnace slags		0.238	0.557	0.736	0.697	0.590	0.712	0.813
Bottom ash and fly ash		0.520	1.029	1.317	1.254	1.083	1.279	1.441
Phosphogypsum		0.661	1.252	1.577	1.506	1.313	1.533	1.710
Bauxite residue		1.830	3.192	3.971	3.798	3.337	3.868	4.323
Cement		-0.019	0.132	0.219	0.199	0.148	0.207	0.257
Scenario number		1	2	3		4	5	6

587
 588 Table 8 shows different index and dose values for 5 types of building materials calculated via
 589 different models described in Table 2. It must be noted that different authors and models use
 590 different background reductions like mentioned in Table 2. In addition in all calculations it is
 591 assumed that both the walls as the floor/ceiling have the same density and thickness. The ACI
 592 calculation is a non-flexible calculation and assumes a density of 2350 kg/m³ and walls of 20
 593 cm thick and is considered as a reference for comparison since this is screening tool prescribed
 594 by the EU-BSS. Looking at a building material with density of 2350 kg/m³ and thickness of 20

595 cm, the index value of the ACI is higher than the index value of I(pd) except for the
596 phosphogypsum composition.

597 In case of building materials lighter than 2350 kg/m³ and thinner than 20 cm building
598 materials, the ACI overestimates the index-value in comparison to I(pd) (scenario 1). In
599 scenario 2 and 3 the building material is thinner than 20cm and heavier than 2350 kg/m³. In
600 scenario 2 solely overestimations by the ACI are observed. In scenario 3, an overestimation by
601 the ACI only occurs in case of bauxite residue and cement. In contrast an underestimation
602 occurs in case of furnace slags, bottom and fly ashes and phosphogypsum. In scenarios 4 and
603 5 the building material is lighter than 2350 kg/m³ and thicker than 20 cm. In scenario 4, the
604 ACI overestimates the index value in comparison I(pd). In scenario 5 an overestimation by the
605 ACI occurs in case of bauxite residue. In contrast an underestimation occurs in case of furnace
606 slags, bottom and fly ashes, phosphogypsum and cement. Looking at building materials
607 heavier than 2350 kg/m³ and thicker than 20 cm, the ACI underestimates the index-value
608 (scenario 6).

609
610 In scenario 1 the ACI underestimates the index-value, it is recommended to use the I(pd) when
611 that ACI index value is above 1. As in scenario 3 and 5 the ACI can over or underestimate the
612 index value it is best to use the I(pd). As the density and thickness parameters of scenario 2
613 and 4 correspond to the parameters of scenario 3 and 4 respectively, it is best to also use the
614 I(pd) for scenario 2 and 4. For scenario 6 the ACI underestimates the index value and from a
615 radioprotection point of view I(pd) is recommended.

616
617 As underestimations by the Mark_{orig} model (corresponds to ACI) in comparison to D(pd)
618 (corresponds to I(pd)) occur in scenario 2, 3, 5 and 6, it is recommended from a
619 radioprotection point of view to use the D(pd) calculation. In other scenarios it is
620 recommended to use the D(pd) calculation when the effective dose approximates 1mSv/y.

621
622 Comparing the D(pd) with EGDA>0% one can see that the D(pd)-dose-values are for all
623 scenarios higher than the ones calculated via EGDA>0% except for scenario 1 and 2, which
624 have a low wall thickness. In both scenarios, the EGDA>0% gives an effective dose which is
625 solely a few μSv/y higher. In scenario 3, 4, 5 and 6 the EGDA>0% gives a dose which is from
626 the order of 10 μSv/y to several 100's μSv/y lower. Therefore, in these scenarios, in case of an
627 effective dose close to 1mSv calculated by D(pd), the authors recommend using a more
628 detailed dose assessment model like EGDA>0% to more accurately assess the dose. It must
629 also be noted that in this comparison the density and thickness of the walls and floor/ceiling
630 are all equal. This can be different in reality and can affect the dose significantly. In addition
631 one has to take into account that a room size larger than 400 cm x 500 cm x 280 cm gives rise
632 to a dose increase like discussed in section 3.2.3. In addition, the presence of windows and
633 doors will also impact this background correction as well as the different sample compositions.

634
635 Regarding the different residues, the AC of a residue can vary according to the input, process
636 parameters, etc. [47,48]. Therefore one cannot draw conclusions from the index and dose
637 values of Table 8 on the usage of these classes of residues as building material but a case by
638 case approach should be performed.

639
640 **4. Conclusion**

641

642 The current study provides a dose calculation assessment based on the original dose
643 calculation of Markkanen with expanded number of gamma lines and higher total gamma
644 intensity. It is shown that working with averaged gamma lines increases the absorbed dose
645 rate in air for ^{238}U and ^{232}Th with 6.1 % and 0.9 % respectively in case of a standard concrete
646 room. In contrast, a decrease of 4.6 % is determined in case of ^{235}U .

647
648 The presence of the build-up increases the absorbed dose rate in air and plays an important
649 role in the final obtained dose received from building materials. In case the build-up is absent,
650 a decrease in absorbed dose rate in air of 52 %, 50 %, 48 % and 67 % for respectively ^{238}U ,
651 ^{232}Th , ^{40}K and ^{235}U is observed. In case of the ISS_{Pelli} model, the use of the Pelliccioni Berger
652 parameters lowered the absorbed dose rate in air with 2.8 % and 2.6 % for respectively ^{238}U
653 and ^{232}Th in comparison with the ISS_{orig} model, which uses the Berger parameters described
654 by Markkanen. Further improvements on the accuracy of the B and consequently the
655 absorbed dose rate in air can be made by working with build-up factors customized towards
656 the chemical composition of the building material with for example a geometric progression
657 approach [49].

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663 The developed EGDA>0% model is complementary to the existing ACI/original Markkanen
664 model and $I(\rho d)/D(\rho d)$ index/dose calculations which prove relevant for the dose assessment
665 within the European legislative framework applicable towards building materials. Due to its
666 simplicity the authors recommend to perform a first screening by using the ACI proposed by
667 the EU-BSS in the case of building materials thinner than 20 cm or lighter than 2350 kg/m³. In
668 the case of a building material thicker than 20 cm or heavier than 2350 kg/m³, the authors
669 propose to use D(ρd) calculation tool of Nuccetelli et al. [2] in case of standard room sizes. In
670 case the resulting dose of this calculation exceeds 1 mSv/y one should perform a more
671 detailed dose assessment. The EGDA>0% model can be used for these specific cases. The
672 EGDA>0% model also allows coping with non-standard room sizes or the presence of doors
673 and windows The model does not consider the dose originated by ^{222}Rn exhalation resulting
674 in an overestimation of the total external gamma dose originating from building materials.

675
676 A sensitivity analysis was performed of the EGDA>0% model. The main factors that contribute
677 to increase the absorbed dose rate in air in comparison to a standard concrete room (Volume
678 of 56 m³; density of 2350kg/m³; wall/floor/ceiling thickness of 20 cm) are

- 679 • Increasing density; in case of 3500 kg/m³ an increase of 9 %, 10 %, 11 % and 2 % for
680 respectively ^{238}U , ^{232}Th , ^{40}K and ^{235}U is observed.
- 681 • Increasing thickness; in case of 80 cm thick walls an increase of 6 %, 7 %, 8 % and 1 %
682 for respectively ^{238}U , ^{232}Th , ^{40}K and ^{235}U is observed.
- 683 • Increasing volume; in case of a room volume of 280 m³ an increase of 5 %, 6 %, 6 %
684 and 2 % for respectively ^{238}U , ^{232}Th , ^{40}K and ^{235}U is observed.

685
686 The main factors that contribute to decrease the absorbed dose rate in air in comparison to a
687 standard concrete room are:

- 688 • Decreasing density; in case of 1000 kg/m³ a decrease of 34 %, 35 %, 38 % and 20 % for
689 respectively ²³⁸U, ²³²Th, ⁴⁰K and ²³⁵U is observed.
- 690 • Decreasing thickness; in case of 5 cm thick walls a decrease of 27 %, 28 %, 29 % and
691 21 % for respectively ²³⁸U, ²³²Th, ⁴⁰K and ²³⁵U is observed.
- 692 • Decreasing volume; in case of a volume of 2.8 m³ a decrease of 4 %, 4 %, 4 % and 3 %
693 for respectively ²³⁸U, ²³²Th, ⁴⁰K and ²³⁵U is observed.
- 694 • Presence of windows or doors; in case of one window of 2 x 2 m in wall 1 a decrease
695 of 4 % for ²³⁸U, ²³²Th, ⁴⁰K and ²³⁵U is observed.
696

697 In addition, the shape of the room can also impact the absorbed dose rate in air. Also the
698 position and size of the window or door in the wall will impact the final absorbed dose rate in
699 air. Larger windows positioned in the middle of the wall lead to a lower absorbed dose rate in
700 air. The implementation of the chemical composition in the model via the attenuation
701 coefficients showed limited effects on the absorbed dose rate in air. For a standard room an
702 increase of 1.4 %, 0.9 % and 2.1 % is observed for ²³⁸U, ²³²Th and ⁴⁰K in case of a FSIP sample
703 composition in comparison to a concrete sample composition. In contrast, a decrease of 6.8%
704 in case of ²³⁵U is observed.

705 Although the Markkanen room model is widely spread and used as a conservative screening
706 tool in European legislation, the uncertainty of the method should be assessed. The expansion
707 proposed here expands the model with validated scientific data but does not take care of the
708 uncertainty. The uncertainty assessment is a topic for further research.
709

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714 References

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