Exposure caused by natural radionuclides in building materials: current practice, regulations and radiation protection standards development

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Abstract

In this paper current radiation protection concepts to limit exposure due to natural radionuclides in building materials and practice of dose modelling and dose assessment are given. Examples of building material evaluations are discussed with regard to approaches for future requirements of radiation protection regulations and standards.

Generally, indoor exposure from building material originates from radionuclides of the natural decay chains U-238, Th-232, U-235, from radon progenies like Rn-222 and Rn-220 as well as the primordial radionuclide K-40. External radiation exposure by gamma radiation and beta particle emissions plus internal exposure due to inhalation of radon progenies causes chronic exposure of the public.

The research in this radiation protection issue provides a state-of-the-art basis for harmonised guidelines and standards to limit the existing public indoor exposure due to the radioactivity of building materials.

Keywords

Natural radiation, building materials, public exposure, existing exposure situations, U-238 decay chain, Th-232 decay chain, K-40

1. Introduction

In most building materials, particularly in those with mineralogical origin, natural radioactivity is evident (e.g. Trevisi et al. 2012). Generally radioactivity in building material originates from the natural decay chains U-238 and Th-232 as well as from the primordial radionuclide K-40. External radiation exposure by gamma radiation and beta particle emissions plus internal exposure due to inhalation of Rn-222 cause chronic exposure of the public. Due to increasing indoor habitation of persons - in average about 80 % of live-time persons stay indoor - external and internal exposure caused by building materials are of increasing importance. Additionally the rising production of building materials using industrial by-products and residual materials from NORM industry necessitate the consideration and regulation of this radiation protection issue in regard to chronically public exposure.

2. Examples of current practice in Europe

In the Czech Republic the 'Draft regulation No 2005-452-CZ' is amending the Regulations of the State Office for Nuclear Safety No 307/2002 Coll. of June, 13th 2002 on Radiation Protection. The Ra-226 activity concentrations of building material used in buildings with habitable rooms shall not exceed 150 Bq/kg. Additionally the activity concentration index *I* is defined in the Regulations of the State Office for Nuclear Safety No 307/2002 Coll. of June, 13th 2002 according to EC RP 112 (1999) and should not exceed 0,5. This criterion corresponds to an annual effective dose of 0.3 mSv and is much more severe than the requirements in the proposed European Basic Safety Standards EC BSS (2011) of 1 mSv per year (I = 1) as in Austria and Poland. In justified cases, I > 0,5 is possible according to the Czech study 'Radiační ochrana' edited by SÚJB (2009). Additional some specific application parameters could be taken into account e.g. the ratio of weight per unit area of the building material to fulfil the requirement.

In Poland the relevant regulation is the Regulation of the Council of Ministers (January, 2nd 2007). Reference is made to construction products and the activity concentrations of Ra-226, K-40 and Th-228 evaluating the activity concentration index $I < 1$ according to EC RP 112 (1999) and in addition the activity concentration of Ra-226 < 200 Bq/kg. Both limits should not exceed by +20 %.

In Finland, in the technical guide 12.2 STUK (2010) reference is made to construction products and activity concentrations of Ra-226, K-40, Th-232, and in specific cases Cs-137 to evaluate the activity concentration index *I* according to EC RP 112 (1999). If appropriate, the radiation dose requirement (1 mSv per year) can be evaluated case-specifically without examination of the activity concentration index of the building material.

3. Existing and notified regulation in Austria

The limits in the Austrian Natural Radiation Sources Ordinance NatRadOrd (2008) and the Austrian standard for radioactivity in building materials ÖNORM S 5200 (2009) are based on the average natural radiation exposure outdoor and the mean activity concentrations in building materials in Austria. Due to the increasing consideration of the exposure due to exhalation of Rn-222 from building materials, the dose caused by inhalation of Rn-222 (progenies) is strictly considered in the Austrian building material standard (ÖNORM S 5200, 2009).

Also the specific application parameters (ε, d) of the building material are considered in the dose assessment.

The radioactivity concentrations requirement for TENORM used in building materials according to the Austrian NORM Ordinance NatRadOrd (2008) is shown in formula (1)

$$I = \frac{a_{Ra-226} - 40 \cdot Bq \cdot kg^{-1}}{40 \cdot Bq \cdot kg^{-1}} + \frac{a_{Th-232} - 25 \cdot Bq \cdot kg^{-1}}{240 \cdot Bq \cdot kg^{-1}} + \frac{a_{K-40} - 370 \cdot Bq \cdot kg^{-1}}{4000 \cdot Bq \cdot kg^{-1}} \quad (1)$$

The relevant Austrian regulations which are notified in the European Union are the amendments to the building materials list "ÖE" of 15. 12. 2002 for the 2nd edition of the building materials list "ÖE" (OIB, 2004) and the amendments to the existing Building Materials List "ÖE" of December, 1st 2004 enacting the third edition of the Building Materials List "ÖE" (OIB, 2006). The relevant Austrian standard is ÖNORM S 5200 (2009) which introduces a more sophisticated formula for the activity concentration index *I*. This requirement also takes the radon emanation factor ε, the building material density ρ and the thickness *d* for the material in use into consideration (formula 2).

$$I = \frac{a_{Ra-226}}{880 \cdot Bq \cdot kg^{-1}} \left(1 + 0{,}07 \cdot \varepsilon \cdot \rho \cdot d\right) + \frac{a_{Th-232}}{530 \cdot Bq \cdot kg^{-1}} + \frac{a_{K-40}}{8800 \cdot Bq \cdot kg^{-1}} \quad (2)$$

4. Future developments

In Annex VII of the proposal for the (revised) European Radiation Protection Directive (EC BSS, 2011) the definition and use of the activity concentration index for the gamma radiation emitted by building materials is appointed. For the purposes of Article 75(2) of the directive proposal, for identified types of building materials, the activity concentrations a_i of natural radionuclides Ra-226, Th-232 and K-40 shall be determined to identify the activity concentration index *I* of the building material:

$$I = \frac{a_{Ra-226}}{300 \cdot Bq \cdot kg^{-1}} + \frac{a_{Th-232}}{200 \cdot Bq \cdot kg^{-1}} + \frac{a_{K-40}}{3000 \cdot Bq \cdot kg^{-1}} \quad (3)$$

where a_{Ra226}, a_{Th232} and a_{K40} are the activity concentrations in Bq/kg of the corresponding radionuclides in the building material.

The activity concentration index is intended as a screening tool for identifying building materials that may be exempted or subject to restrictions. For this purpose the activity concentration index may be used for the classification of the materials into four classes, leading to two categories of building materials A or B and two classes of use {1} or {2} (Tab. 1).

Table 1 Proposed classification of radioactivity in building material according to EC BSS (2011)

	Category (corresponding default dose)	
Use	A (≤ 1 mSv)	B (> 1 mSv)
{1} materials used in bulk amounts	I ≤ 1	I > 1
{2} superficial and other materials with restricted use	I ≤ 6	I > 6

The allocation of materials into A or B and into {1} or {2} according to their use should be based on national building codes.

Where appropriate, actual doses for comparison with the reference level should be assessed using more elaborate models, which may also take into account the background outdoor external exposure from local prevailing activity concentrations in the undisturbed earth's crust.

On the occasion of the CEN Technical Committee 351 ‘Construction Products - Assessment of release of dangerous substances’, Working Group 3 ‘Radiation from construction products’, a task group was established (TG 3.2 ‘Dose modeling’) which should identify a more realistic model for the assessment of the external exposure due to radionuclides in building materials. The application of the activity index according to the concept of the RP 112 (1999) and used as basis for Annex VII of the proposed EC BSS (2011) is limited explicitly as "screening tool" for the identification of possibly relevant materials.

Therefore, formula (3) according to Annex VII of the proposed BSS Directive could be amended by a weighting factor taking into account the density ρ and the thickness d of the finally built-in material (CEPMC, 2011).

$$I = \left(\frac{a_{Ra-226}}{300 \cdot Bq \cdot kg^{-1}} + \frac{a_{Th-232}}{200 \cdot Bq \cdot kg^{-1}} + \frac{a_{K-40}}{3000 \cdot Bq \cdot kg^{-1}} \right) \frac{d \cdot \rho}{470 \cdot kg \cdot m^{-2}} \quad (4)$$

a_i = activity concentrations of the radionuclide i in Bq/kg

d = thickness of the wall, superficial or other material in m

ρ = gross dry density of the material in kg/m³

470 kg/m² is the weight per unit area of the model room in EC RP 112 (1999)

(Table 3, concrete, thickness 0,2 m, density 2350 kg/m³)

If the index evaluation should be applied to other materials than concrete, supplementary the raw density has to be considered. If necessary, the decrease of the cosmic ray shielding could be taken into account. Also a limit of chargeable component thickness and/or a limit of the maximum radionuclide concentration for Ra-226 lower than 200 Bq/kg is reasonable.

5. Results of building material radioactivity measurements

In Austria there is a long experience in checking and limiting the natural radioactivity of building materials. In 1995 an Austrian standard for the evaluation of building material radioactivity has been established. Since 2002 this standard is implemented obligatory into the Austrian federal country’s construction product codes. This implementation had been done by including technical annexes to the construction product codes (OIB, 2004, OIB, 2006). The Austrian construction product codes are notified by the EU member states.

In Tables 2a and 2b, examples of building material radioactivity measurements done since 30 years at the Low-level Counting Laboratory Arsenal, Vienna, are given (84 samples).

The conservative application parameter values d * ρ for the index (4) calculation in Tables 2a and 2b are 0,3 m * 700 kg/m³ for bricks and 0,2 * 2350 kg/m³ for all other materials.

Table 2a **Results of activity concentration measurements of 42 building material samples and index *I* evaluation: (2) - ÖNORM S 5200 (2009), (3) - EC BSS (2011), (4) - application formula**

Sample code	Activity concentration Bq/kg			Index I (formula)		
	^{40}K	^{226}Ra	^{232}Th	(2)	(3)	(4)
2-R	631	24	41	0,31	0,50	0,22
4-R	661	33	51	0,39	0,59	0,26
9-R	547	56	62	0,51	0,68	0,30
6-R	569	64	66	0,53	0,73	0,33
1-Z	667	30	38	0,33	0,51	0,23
3-Z	755	43	46	0,46	0,63	0,28
8-Z	571	54	64	0,50	0,69	0,31
10-Z	754	85	62	0,69	0,84	0,38
5-Z	505	47	61	0,41	0,63	0,28
7-Z	437	35	56	0,38	0,54	0,24
67-S	417	14	25	0,24	0,31	0,31
11-Z	890	46	60	0,42	0,75	0,34
68-S	0	6	19	0,14	0,11	0,11
69-S	9	4	11	0,12	0,07	0,07
13-Z	632	42	43	0,36	0,57	0,25
14-Z	597	45	47	0,35	0,58	0,26
16-Z	589	41	48	0,40	0,57	0,26
15-Z	447	50	53	0,43	0,58	0,26
12-Z	609	44	34	0,44	0,52	0,23
18-Z	690	39	47	0,41	0,60	0,27
17-Z	579	32	38	0,35	0,49	0,22
19-Z	663	39	49	0,37	0,60	0,27
64-G	0	770	8	4,65	2,60	2,60
20-Z	586	37	42	0,37	0,53	0,24
21-Z	667	54	65	0,47	0,73	0,32
22-Z	523	66	53	0,54	0,66	0,29
25-R	740	31	44	0,39	0,57	0,25
26-R	590	28	33	0,33	0,46	0,20
28-R	100	76	140	0,52	0,99	0,44
23-Z	588	37	46	0,30	0,55	0,25
24-Z	495	61	61	0,50	0,67	0,30
27-Z	580	51	65	0,45	0,69	0,31
30-Z	736	20	29	0,29	0,46	0,20
29-Z	650	91	55	0,88	0,80	0,36
70-N	1130	60	109	0,59	1,12	1,12
78-N	931	109	143	1,00	1,39	1,39
79-N	1100	49	61	0,52	0,84	0,84
80-N	517	32	36	0,35	0,46	0,46
81-N	269	8	11	0,18	0,17	0,17
82-N	1110	99	93	0,81	1,16	1,16
83-N	1330	108	89	1,04	1,25	1,25
84-N	1390	26	34	0,52	0,72	0,72

Table 2b **Results of activity concentration measurements of 42 building material samples and index *I* evaluation: (2) - ÖNORM S 5200 (2009), (3) - EC BSS (2011), (4) - application formula**

Sample code	Activity concentration Bq/kg			Index I (formula)		
	^{40}K	^{226}Ra	^{232}Th	(2)	(3)	(4)
71-S	149	6	5	0,17	0,10	0,10
72-S	193	8	8	0,10	0,13	0,13
73-S	187	13	11	0,14	0,16	0,16
74-S	400	14	12	0,26	0,24	0,24
75-S	114	19	5	0,28	0,13	0,13
76-S	46	47	3	0,40	0,18	0,18
77-S	31	23	3	0,25	0,10	0,10
31-Z	750	39	43	0,43	0,60	0,27
33-Z	650	91	55	0,67	0,80	0,36
34-Z	652	63	45	0,55	0,65	0,29
32-Z	612	74	43	0,63	0,67	0,30
63-B	1000	86	85	0,75	1,04	1,04
58-Bt	513	27	30	0,28	0,41	0,41
59-Bt	510	37	36	0,34	0,47	0,47
60-Bt	586	40	32	0,55	0,49	0,49
61-S	712	24	35	0,25	0,49	0,49
62-S	588	26	35	0,32	0,46	0,46
65-F	708	260	128	1,70	1,74	1,74
66-N	1070	156	17	1,14	0,96	0,96
44-Z	740	53	51	0,41	0,68	0,30
41-Z	720	92	67	0,79	0,88	0,39
39-Z	850	48	53	0,40	0,71	0,32
38-Z	790	45	45	0,42	0,64	0,29
36-Z	730	37	49	0,57	0,61	0,27
42-Z	670	42	57	0,39	0,65	0,29
37-Z	810	41	56	0,45	0,69	0,31
40-Z	840	41	52	0,51	0,68	0,30
43-Z	870	47	58	0,39	0,74	0,33
35-Z	790	53	62	0,61	0,75	0,34
45-Z	895	57	15	0,46	0,56	0,25
46-Z	728	118	20	0,80	0,74	0,33
47-Z	710	61	69	0,45	0,79	0,35
48-Z	820	116	81	0,78	1,07	0,48
49-Z	920	96	62	0,68	0,94	0,42
50-Z	815	97	59	0,75	0,89	0,40
57-Z	1110	110	130	0,84	1,39	0,62
55-Z	770	54	54	0,46	0,71	0,32
53-Z	703	53	51	0,47	0,67	0,30
54-Z	745	55	52	0,57	0,69	0,31
56-Z	810	61	54	0,52	0,74	0,33
51-Z	819	91	135	0,73	1,25	0,56
52-Z	880	71	112	0,60	1,09	0,49

Figure 1 shows the sum frequencies of the different radioactivity index of building material samples evaluated by the formulas: (2) - ÖNORM S 5200 (2009), (3) - EC BSS (2011), (4) - application formula. The graph is a sum frequency box plot with 10 %, 25 %, 50 %, 75 %, 90 % quantiles and single data points out of the sum frequency range 10 % - 90 %. The sum frequencies of the three indexes are nearly log-normal distributed. Highest index values originate from the EU BSS formula (3), lowest from the application parameters formula (4) (Fig. 1).

The detailed distributions of the index values are shown in Fig. 2 and 3 according to formula (3) – EU BSS and application formula (4), respectively. The index values of sand samples are in the lower part, the brick samples in the medium and the natural stone samples in the higher part of the index range.
By using formula (3) about 12 % of building material give an index value higher than 1 whereas with application parameters formula (4) only 8 % of the material shows index values higher than 1. This fact indicates marginal differences for identification of building materials above radioactivity limits between formula (3), EU BSS, and (4), application parameters.

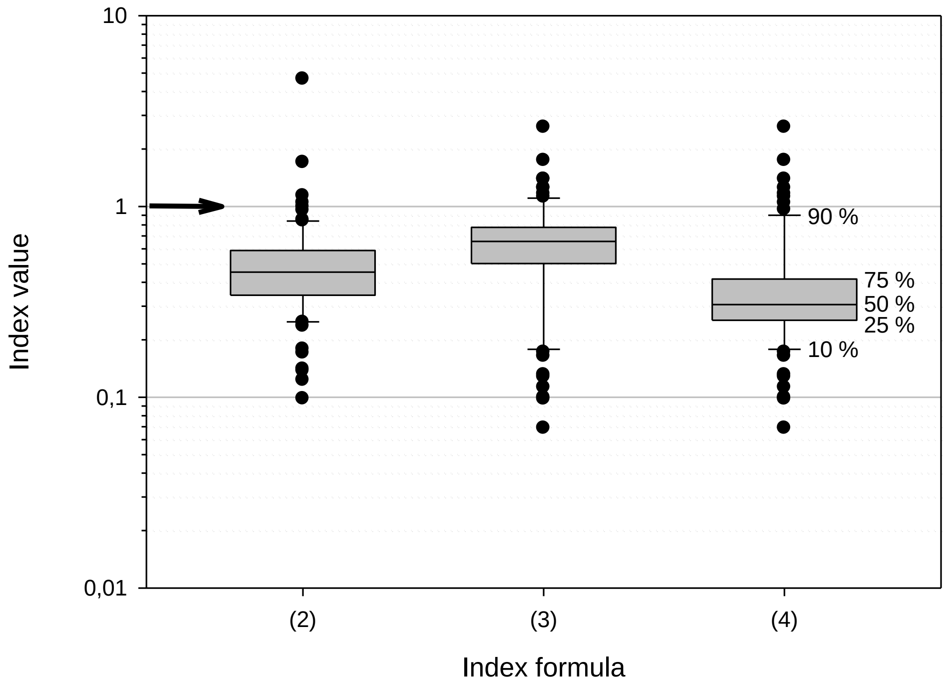

Fig. 1 **Sum frequency of radioactivity index of 84 building material samples from different evaluation formula: (2) - ÖNORM S 5200 (2009), (3) - EC BSS (2011), (4) - application formula** (*box plot:* 10 %, 25 %, 50 %, 75 %, 90 %, and single data points)

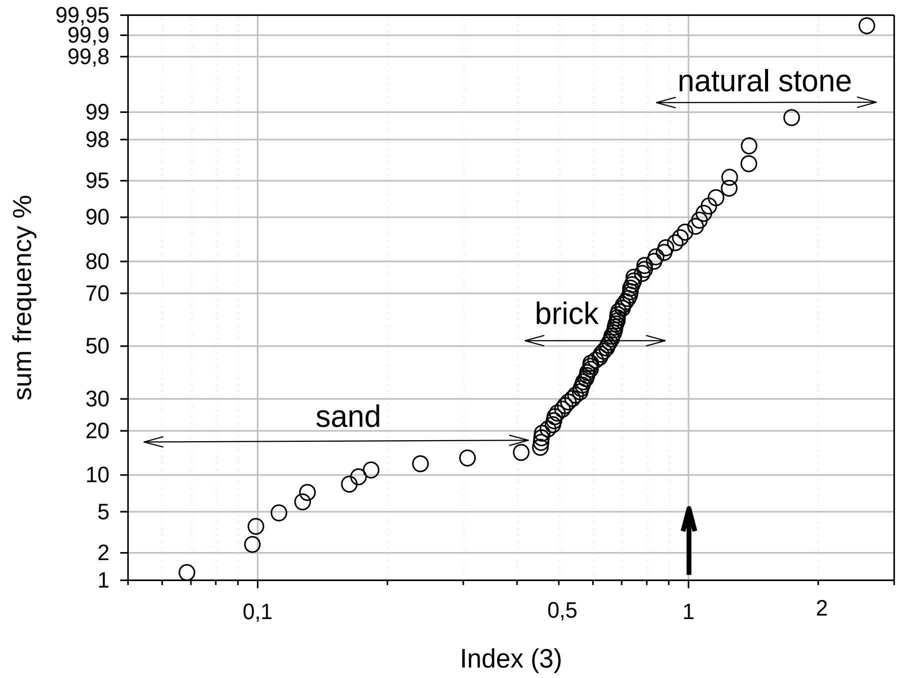

Fig. 2 **Sum frequency distribution of radioactivity index of 84 building material samples from evaluation formula (3) - EC BSS (2011)**

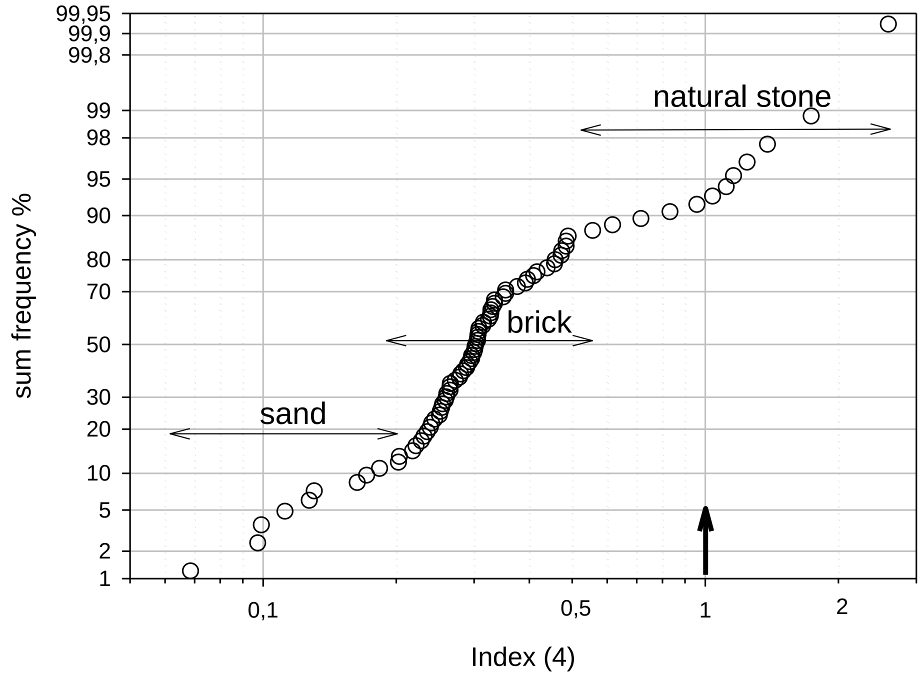

Fig. 3 **Sum frequency distribution of radioactivity index of 84 building material samples from evaluation formula (4) – application parameters formula**

6. European and international standards development

In consequence, a common dose assessment model for use by EU Member States, whether within the context of the EU Construction Products Directive or the revised EC BSS (2011), would be extremely useful and could contribute to harmonization and standardization in this area. To this end, TC351/WG3/TG32 proposes the development of a CEN Technical Report which allows all the issues to be explored and explained, including the shortcomings of Member States' 'activity concentration indices' and the EC RP 112 (1999), 'activity concentration index'.

For dose modelling some decisions have to be taken for parameter values:

- The 'typical level' for outdoor exposure
  (The reference limit 1 mSv per year for exposure due to building material's radioactivity is defined as the "excess to typical outdoor exposure". Outdoor exposure rates vary locally.)
- The geometry of the standard room to be used in the indoor dose model
- The indoor occupancy time
- The shielding effect of building materials for cosmic radiation
- Assumptions on other materials in the building than the examined
- Factor for converting the ambient dose equivalent in air to the effective dose

7. Conclusions

The rough differentiation of materials in the proposed EC BSS (2011) "Materials used in bulk amounts" of index ≤ 1 and "Superficial" with index ≤ 6 (Tab. 1) is ineffective to limit consequently the external exposure due to natural radionuclides (U-238 and Th-232 decay chain, K-40) in building materials. To assess the exposure realistically, the actual application parameter values of thickness and bulk density (specific mass) of the building material should be considered in the evaluation procedure.

The threshold dose in EC BSS (2011) is based on the model room assumptions for walls, floor and ceiling made of concrete with 0,20 m of thickness and 2350 kg/m³ of density according to EC RP 112 (1999). For the evaluation of the activity index of other construction materials a weighting factor for different material thicknesses and densities should be added. The index limit (I ≤ 6) for "superficial and other materials with restricted use" (Tab. 1) is not effected significantly by these parameters.

To ensure the conformity of construction products with radiation protection requirements, it is recommended to appoint only the dose limit itself and transfer the dose modeling, limit assessment and radioactivity measurement to an harmonised technical standard. This could be done for example in the European Union within the CEN TC 351 Working Group 3, Task Groups 3.1 'Gamma-ray measurement of building materials' and 3.2 'Dose modeling'.

In consequence, a harmonised dose assessment model for the external exposure due to natural radionuclides in building materials would be extremely useful. A common standard could diminish the number of trade disputes and contribute to the international harmonization in radiation protection. In the European Union, the CEN Technical Committee 351 ‘Construction Products - Assessment of release of dangerous substances’, Working Group 3 ‘Radiation from construction products’, Task Group 32 ’Dose modeling’ proposes the development of a CEN Technical Report which allows all the issues to be included especially realistic building material application parameter values. The proposed CEN Technical Report could act as recommendation to European regulators and international radiation protection authorities to adopt a common and harmonised assessment to limit the exposure to the public due to natural radionuclides in building materials.

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