Radioactivity in Building Materials: a first Overview of the European Scenario

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Abstract. With a wide research into the national and international literature an inventory was created of building materials in Europe, characterised on the basis of activity concentration of the main natural radionuclides (^{226}Ra, ^{232}Th and ^{40}K). Materials of natural origin and containing industrial by-products were both accounted for. The inventory allowed us to calculate the activity concentration index I - suggested by a European technical guidance document - for many building materials in Europe. A first identification of materials was thus made, which could be subject to controls or restrictions as for movement and/or use if the index were to be adopted by the European legislation.

KEYWORDS: building material, activity concentration, natural radioactivity, European legislation.

1. Introduction

In the last decades building materials, both of natural origin and containing industrial by-products, have been shown to significantly contribute to the exposure of the population to natural radioactivity. As a matter of fact, neither the absorbed dose rate in air due to gamma radiation nor the radon activity concentration are negligible in closed environments.

In 1999 the technical guidance document *Radiation Protection 112* [1] suggested the calculation of an index I for limiting the natural radionuclide activity concentration in building materials, only considering gamma radiation. The same document provided a grid of limit values for this index as a function of the dose criterion adopted and the material's intended use (structural or superficial).

In the late 90's of last century an inventory of Italian building materials (over one thousand) was organised [2]. It was based on original data from the authors and data from the literature. Materials were characterised in terms of activity concentration of the main natural radionuclides (^{226}Ra, ^{232}Th and ^{40}K). The possible consequences of imposing restrictions on their marketing and use were also evaluated [2].

Recently, this investigation was extended to include European building materials, considering the possible adoption of the index I [1] by the European Commission (EC) in a new comprehensive EURATOM directive, currently under discussion in the Expert Group, referred to in art. 31 of the EURATOM Treaty. If the index I is adopted, the European Council Directive related to construction products, issued in 1989 [3], will finally find a practical application. It provides that buildings be designed and constructed in such a manner that they pose no risk to the health of occupants or neighbours, resulting from emissions of hazardous radiation. But as it gives no indication of numerical parameters to be met, it has been widely judged unenforceable up to now.

A review of the literature regarding European building materials had already been made in 1996, under a contract of the EC [4]. At that time, however, the Member States (MS) were only 15 – 27 today – and data were collected for 10 out of the 15 MS and other non MS. Moreover, the European technical guidance [1] was published later on, so that in the cited publication [4] no analysis with limitation criteria, such as the

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application of the index I, had been attempted. Finally, even if the majority of data on natural radionuclides in building materials was published in the 80’s and 90’s of last century, with a thorough research it was possible not only to enrich the data base, but to update it as well.

2. The European Technical Guidance

The European technical guidance [1] is a reference document for the European Commission in view of future legislative initiatives. It suggests to base radioactivity controls of building materials on the choice of a dose criterion for controls and an exemption level.

Dose criterion for controls. The dose criterion for controls should be established considering the overall national circumstances. Within the European Union (EU) effective doses exceeding 1 mSv y^{-1} should be taken into account from the radiation protection point of view. Higher doses should be accepted only in highly specific circumstances where materials are locally used. However, if deemed appropriate and not impractical, controls can be based on a lower dose criterion. In conclusion, it is recommended to choose dose criteria nationally in the range 0.3 - 1 mSv y^{-1} of excess gamma dose to that received outdoors.

Exemption level. Building materials should be exempted from all restrictions, concerning their radioactivity, if the excess gamma radiation originated from them increases the annual effective dose of a member of the public by 0.3 mSv at the most. This excess dose has to be calculated by subtracting the effective dose received outdoors, estimated in 50 nGy h^{-1} for the European Union countries. This average value – weighed on the population – had been evaluated from the national average data of the UNSCEAR 1993 report [5]. Since several radionuclides contribute to the dose, in order to assess whether the dose criterion is met, a level of investigation was suggested in the form of an activity concentration index I:

$$I = \frac{C_{Ra-226}}{300 Bqkg^{-1}} + \frac{C_{Th-232}}{200 Bqkg^{-1}} + \frac{C_{K-40}}{3000 Bqkg^{-1}} \tag{1}$$

where C_x is the activity concentration of radionuclide x in the sample considered and ^{226}Ra is used as reference for the parent ^{238}U. The limit values chosen for the index I depend on the dose criterion adopted and the use of the material (see Table 1).

Table 1: Limit values for the index I according to the dose criterion adopted

Dose criterion (a)	0.3 mSv y^{-1}	1 mSv y^{-1}
Materials used in bulk amounts	$I \leq 0.5$	$I \leq 1$
Superficial and other materials with restricted use	$I \leq 2$	$I \leq 6$

$^{(a)}$ effective dose rate excess to that received outdoors

3. The Inventory of European Building Materials

Activity concentration data of building materials were collected through a large review of the national and international literature [4,6-63] concerning building materials, which are used in European MS, but not necessarily produced in the country where they are used. The exact number of samples corresponding to the measurements in the database could not be assessed, because in some publications the number of samples is not specified (this was indicated as = 1 in the database). Therefore 8992, the total number of samples assessed, is certainly an underestimation of the actual number. The width of activity concentration ranges is also underestimated, because in some publications only the average values are available.

The data base does not claim to offer a representative picture for Europe, as the investigations were not statistically representative in most countries. Moreover, for radiation protection purposes, measurements are generally made on materials that are presumably more active, therefore the values published are probably an overestimation of the actual situation. Finally, many of these measurements were taken in the nineteen eighties and nineteen nineties and is undeniable that the measurements made in those years were less accurate, often because they did not account for radioactive disequilibrium in the ^{238}U chain, coincidence-summing effect, self-absorption in materials with density different from 1, etc. [64-66]. Also for this reason, the inventory considers ^{238}U activity concentrations equivalent to ^{226}Ra ones.

This paper presents a first analysis of data for 22 Member States, on at least 6617 samples of materials used in the construction industry in bulk quantity, namely, brick, concrete, cement, natural gypsum and phosphogypsum. The potential implementation of controls on, or restrictions on the movement and use of, these common materials would have a major economic impact.

The index I [1] was calculated using average, minimum and maximum activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K. The results are presented in Table 2 and 3. The index I could not be calculated for data from Belgium, because ^{40}K activity concentration is missing, but the Belgian data were used for elaborating Table 4 and reported in Fig.1 and 2. The collection has not been concluded and therefore more data will be probably available in a near future (more countries and materials).

In Table 2, a high variability of I_{max} and I_{min} for each type of material in the same country can generally be observed. The very high values of I_{min} for bricks and of I_{max} for concrete in Sweden [34], are most likely due to the use of alum shale. In Danish concrete, too, some samples come from Sweden and contain alum shale [60]. The low variability – and probably the high value – of I_{min} and I_{max} in the brick of Luxembourg is due to the very limited statistics (only 2 samples). Excluding Sweden and Luxembourg, no other I_{mean} for any material is higher than 1, whereas it is less than 0.5 for only a few materials. This means that the goal of not exceeding 1 mSv y^{-1} seems easily achievable, whereas that of not exceeding 0.3 mSv y^{-1} could prefigure a situation difficult to manage. In conclusion, from this first analysis it seems that at the European level the choice of a dose criterion in the range 0.3 - 1 mSv y^{-1} would be feasible, while nationally the choices might differ and some countries could aim at more ambitious goals.

As regards the samples of concrete, it must be noticed that different approaches have been used to assess their activity concentration: sometimes actual samples of concrete were measured, sometimes only ballast, sometimes the different components (cement, sand, gravel, etc.), making assumptions on its composition to obtain the final assessment.

Some cement samples contain fly ash [11, 32, 53, 58] and other samples may as well do so, even if it is not specified. Cement deserves further consideration, because in buildings it is always used as a component of concrete (structural material) or plaster, mortar and other superficial materials: in all cases the index I – calculated without a reduction factor – is overestimated and for superficial material it should be compared with the relevant values of Table 1.

In Table 3 the data available are much less, particularly for phosphogypsum. As expected, the resulting I values are generally very low – even for I_{max} – for natural gypsum, and high – even for I_{min} – for phosphogypsum.

Table 4 reports averages and ranges of activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K for brick, concrete, cement and natural gypsum, for the 22 Member States. These values are compared with the concentrations of the same radionuclides in European soil from the 2000 UNSCEAR report [67]. They were calculated on the basis of UNSCEAR national averages and are not statistically representative. The average ^{226}Ra and ^{40}K activity concentrations of brick are about 40% and 30%, respectively, higher than the average concentrations in European soil. Average ^{232}Th is about 60% higher. For concrete, average concentrations of ^{232}Th and ^{40}K are similar to those of soil, but average ^{226}Ra is almost 80% higher. Cement differs for a low content of ^{40}K, an activity concentration of ^{232}Th comparable with the average content in European soil and a ^{226}Ra average activity concentration 40% higher. Natural gypsum, as already underlined, has a low level of radioactivity. Lastly, a wide variability of the content of natural radionuclides – shown by the ranges – is found in both building materials and soil.

Looking again at brick data, it can be seen that the average concentrations and the ranges of ^{226}Ra and ^{232}Th are almost equal, but a more detailed examination of data from individual countries reveals a more complex situation. In Fig. 1 a comparison is drawn of the content of ^{226}Ra and ^{232}Th in brick in the 22 European MS. In 10 out of the 22 (Austria, Bulgaria, Czech Republic, Luxembourg, The Netherlands, Poland, Romania, Slovenia, Spain and Sweden) ^{232}Th average concentration is higher than the ^{226}Ra one. It means that the study and research into the ^{232}Th chain, currently considered of secondary importance, should instead be deepened [68].

A similar analysis was done for cement data and can be seen in Fig. 2. In this case, the average activity concentration of ^{232}Th is higher than that of ^{226}Ra in 4 MS (Belgium, Italy, The Netherlands, Sweden), whereas in Table 4, the value averaged over all the available data shows that ^{226}Ra activity concentration is higher by more than 40% .

Table 2: Index I calculated with average, maximum and minimum activity concentration of ^{226}Ra, ^{232}Th and ^{40}K for brick, concrete and cement. The values I > 1 are shown in bold, those <0.5 in italics.

	BRICK				CONCRETE				CEMENT			
	N (a)	I_{mean}	I_{max}	I_{min}	N (a)	I_{mean}	I_{max}	I_{min}	N (a)	I_{mean}	I_{max}	I_{min}
Austria	32	0.56	**1.09**	*0.32*	1	*0.17*	*0.48*	*0.04*	18	*0.23*	*0.39*	*0.12*
Belgium	61				8				22			
Bulgaria	1	0.56			2	*0.35*	*0.48*	*0.22*	1	*0.25*		
Czech Republic	488	0.60	0.60	0.59	491	*0.40*			496	*0.33*		
Denmark	83	*0.34*	0.52	*0.16*	121	0.85	**2.90**	*0.19*	6	*0.16*	*0.25*	*0.06*
Finland	42	0.65	0.91	*0.39*	294	0.57	0.65	*0.46*	11	*0.32*	0.67	*0.15*
Germany	135	0.55	1.00	*0.17*	75	0.67	**1.02**	*0.37*	23	0.71	**1.68**	*0.27*
Greece	32	0.59	0.75	*0.29*	49	*0.19*	*0.40*	*0.10*	117	*0.48*	0.92	*0.13*
Hungary	176	0.61	**1.06**	*0.41*	97	*0.28*	*0.37*	*0.17*	400	*0.28*	0.60	*0.12*
Ireland	14	*0.45*	0.81	*0.15*	8	*0.23*	0.54	*0.10*	3	*0.30*	*0.39*	*0.19*
Italy	192	*0.49*	0.98	*0.11*	20	*0.26*	*0.34*	*0.20*	200	0.57	**1.79**	*0.13*
Lithuania	1	0.54			1	*0.33*						
Luxembourg	2	**1.21**	**1.28**	**1.13**	2	0.80	0.82	0.79				
The Netherlands	70	0.50	0.95	*0.15*	55	*0.36*	**1.33**	*0.13*	17	0.62	0.96	*0.26*
Poland	6	*0.32*	0.51	*0.13*	678	0.97	**1.64**	0.56	344	0.69	**1.41**	*0.33*
Portugal	10	0.73			11	0.54			7	*0.32*		
Romania	75	0.55	**1.14**	*0.16*	133	0.67	**1.07**	*0.19*	55	*0.39*	1.00	*0.08*
Slovakia	1	0.62	0.84	*0.36*	41	*0.38*	0.57	*0.16*	6	*0.28*	*0.33*	*0.22*
Slovenia	2	0.93	**1.11**	0.74	3	0.56	**1.37**	*0.16*				
Spain	1	0.83							2	*0.45*	*0.48*	*0.43*
Sweden	33	**1.27**	**1.28**	**1.26**	458	**1.32**	**4.93**	*0.40*	26	0.52	0.61	*0.41*
United Kingdom	80	*0.46*	**1.04**	*0.04*	17	0.54	0.58	*0.45*	6	*0.22*		
TOTAL	1537				2565				1760			

(a) N = number of samples

Table 3: Index I calculated with average, maximum and minimum activity concentration of ^{226}Ra, ^{232}Th and ^{40}K for gypsum and phosphogypsum. The values I > 1 are shown in bold, those <0.5 in italics.

	GYPSUM				PHOSPHOGYPSUM			
	N (a)	I_{mean}	I_{max}	I_{min}	N (a)	I_{mean}	I_{max}	I_{min}
Austria								
Belgium	18				30			
Bulgaria	18	*0.07*	*0.15*	*0.04*	2	0.78	**1.38**	*0.19*
Czech Republic	34	*0.18*			22	0.57		
Denmark	12	*0.05*						
Finland	1	*0.04*			17	**1.14**	**3.37**	*0.10*
Germany	1		0.80	*0.02*	2	**1.97**		
Greece	8	*0.08*	*0.11*	*0.05*	4	**2.07**	**2.20**	**1.83**
Hungary	125	*0.06*	*0.19*	*0.01*				
Ireland	10	*0.14*	*0.18*	*0.03*				
Italy	20	*0.09*	*0.14*	*0.04*				
Lithuania								
Luxembourg								
The Netherlands	23	*0.08*	*0.21*	*0.04*	20	0.88	**1.58**	*0.34*
Poland	105	*0.28*	*0.46*	*0.10*	28	1.00	**1.43**	*0.25*
Portugal								
Romania	15	*0.27*	*0.40*	*0.14*	73	**1.94**	**2.49**	**1.15**
Slovakia								
Slovenia					1	**1.73**		
Spain	1	*0.13*						
Sweden	1	*0.02*						
United Kingdom	73	*0.16*			91	**3.60**	**5.00**	**2.20**
TOTAL	465				290			

(a) N = number of samples

Table 4: ^{226}Ra, ^{232}Th and ^{40}K activity concentrations in European brick, concrete, cement, natural gypsum and soil: average values and typical ranges

Building material	Number of samples	Activity concentration (Bq kg^{-1})		
		^{226}Ra	^{232}Th	^{40}K
Brick	1537	48 (2 – 200)	52 (1 – 200)	619 (12 – 2000)
Concrete	2565	62 (4 – 2620)	36 (1 – 463)	431 (16 – 3120)
Cement	1760	49 (4 – 330)	34 (1 – 240)	226 (1 – 846)
Natural gypsum	465	14 (1 – 70)	8 (1 – 100)	97 (3 – 279)
Soil [67]				
Northern Europe		36 (6 – 310)	32 (5 – 94)	640 (140 – 1150)
Western Europe		37 (5 – 900)	32 (3 – 180)	430 (0 – 3200)
Eastern Europe		33 (1 – 210)	31 (2 – 160)	452 (40 – 1400)
Southern Europe		36 (0 – 250)	35 (1 – 210)	433 (0 – 1650)
European average		35 (0 – 900)	32 (1 – 210)	485 (0 – 3200)

Figure 1: Mean activity concentration of ^{226}Ra and ^{232}Th in bricks of 22 Member States

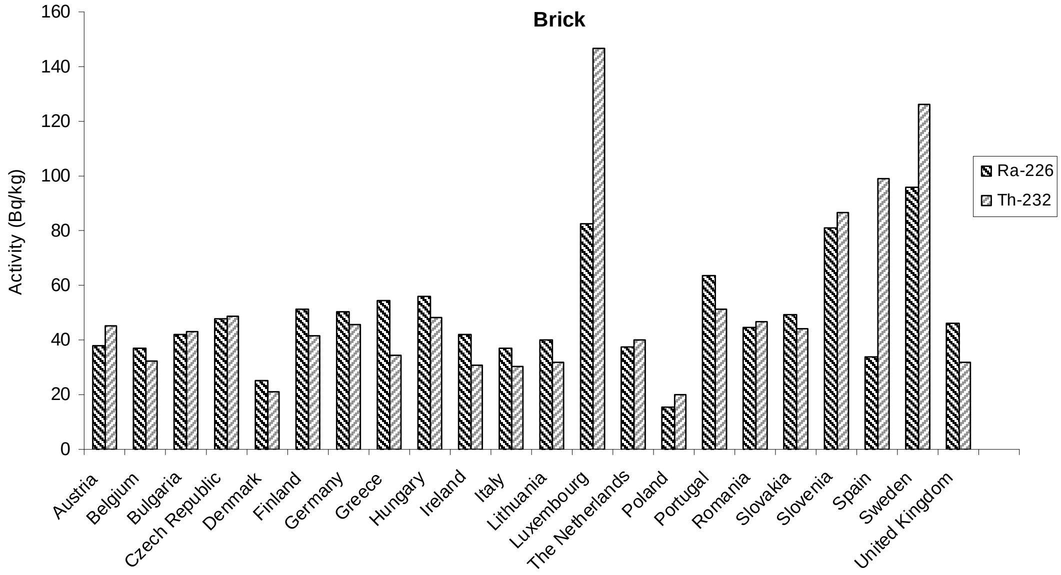
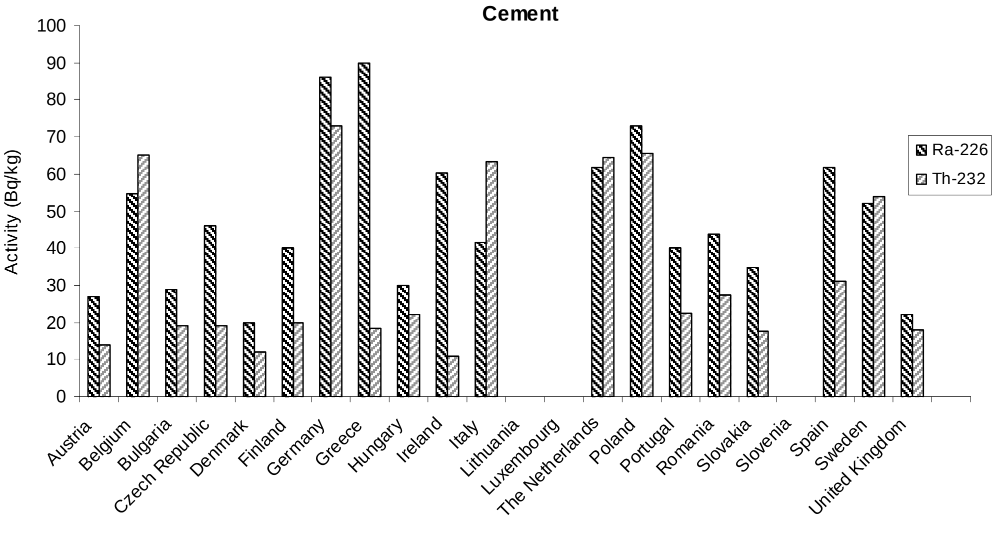


Figure 2: Mean activity concentration of ^{226}Ra and ^{232}Th in cements of 19 Member States



4. Conclusions and future prospects

As specified above, the database is still under construction and a major effort will be made to feed it with as much data as possible, counting on the collaboration of colleagues that will provide us with further data. Once completed, the maximum effective dose for European citizens indoors due to gamma radiation from main building materials will be assessed, at least as a first approximation. As a point of fact, the average exposure cannot be assessed, because, as stated above, these data are not statistically representative of European MS.

From a radiation protection point of view a comment should be made. The dose criterion considers only effective dose due to gamma radiation, but the population at home is also subject to exposure to

^{222}Rn from soil, water, drinking water and the material itself, sometimes to beta radiation from superficial materials, and even to ^{137}Cs in some types of wood, etc. Therefore the population exposure indoors is actually much higher than the value assessed with this dose criteria.

The analysis presented in this paper is a first attempt to discuss the data of our inventory and only five materials have been analysed. In a near future a more complete discussion will be published, also considering natural stones and superficial materials. As regards natural stones a tentative grouping will be made, classifying stones by their geological origin. Moreover, if enough data were available, we will also assess the radiation protection consequences of the potential use of by-products of industrial origin in building materials.

Finally, the activity concentration of ^{232}Th, often higher than that of ^{226}Ra, in building materials shows the need of improving research into the health effects of the ^{232}Th chain, in particular of thoron concentration indoors.

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