Natural radioactivity in building materials in the European Union: a database and an estimate of their radiological significance

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Content

- Database of building material activity concentrations
  - distribution of data in MS
  - distribution of bulk material/product data in MS
    - ✓ bricks
    - ✓ concrete
    - ✓ phosphogypsum
  - Comparison of EU data with EU soil
  - Analisys of bulk materials/products exceeding
    - RP112 criteria
    - Ö-NORM 2009 and SI 5098 criteria
  - Superficial materials
  - Conclusions

Database information

Database contains data for 24 out of the 27 MS – no data for Estonia, Latvia and Malta.

For some MS information was not available for all types of building materials, therefore number of MS and samples involved are different for each material.

Several categories of building materials:

- products used in bulk amounts (bricks, concrete) - about 4400 samples;
- materials (cements, gypsum) used in bulk amounts and superficial application - about 2500 samples;
- natural stones used both as bulk and superficial products - about 900 samples;
- phosphogypsum - about 290 samples;
- industrial by-products, typically fly and bottom ashes - about 1300 samples;
- others such as wood, tiles, etc. - about 500 samples.

Database information (cont.)

- Analysed the first four categories (bulk products, superficial products, natural stones, phosphogypsum) - at least 8132 samples
  - they are largely employed in the construction industry and their contribution to the indoor gamma dose may be relevant.

- Measurements were generally made on materials that were presumed to be more active (radiation protection purposes), therefore the values published are probably an overestimate of the actual situation.

- Notwithstanding the large number of data collected,
  - the numbers of samples for each MS are low in some cases and quite different;
  - a high variability of activity concentrations can generally be observed
    - ✓ ranges show from two to three orders of magnitude between the minimum and maximum values
  - the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K are not available for all the samples
    - ✓ in these cases the radiological significance of building products cannot be analysed by means of indexes.

Sets of data collected and used for each MS

Country	# of samples	# of samples analysed	Country	# of samples	# of samples analysed
Austria	105	95	Italy	1112	738
Belgium	218	214	Lithuania	2	2
Bulgaria	42	38	Luxembourg	89	64
Cyprus	55	55	The Netherlands	219	190
Czech Republic	1531	1531	Poland	1331	1173
Denmark	307	223	Portugal	78	62
Finland	439	366	Romania	737	378
France	44	44	Slovakia	60	57
Germany	299	257	Slovenia	6	6
Greece	1032	572	Spain	423	317
Hungary	849	810	Sweden	625	625
Ireland	35	35	United Kingdom	284	280

24 EU-Member States:

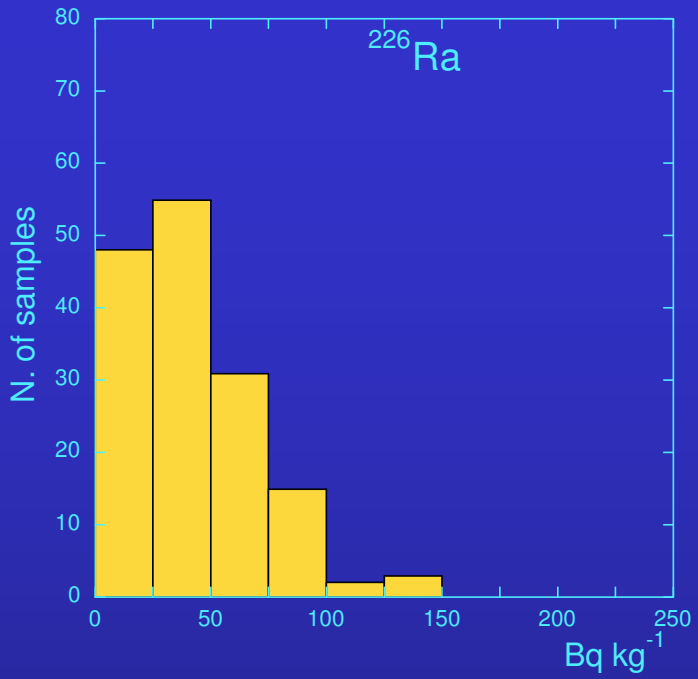
Total # of samples = 9922

Total # of samples analysed = 8132

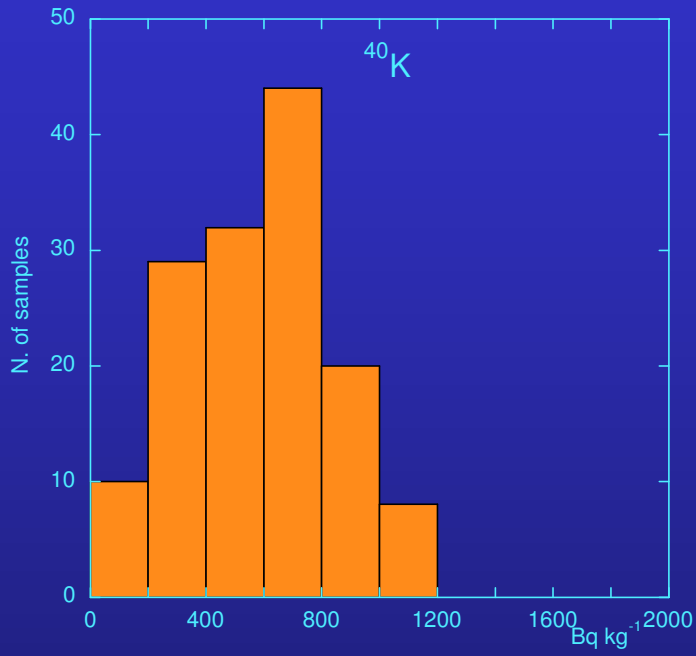
Bricks: activity concentration database (at least 1676 samples)

Country (24 MS)	N of samples	^{226}Ra (Bq kg^{-1})			^{232}Th (Bq kg^{-1})			^{40}K (Bq kg^{-1})		
		average	min	max	average	min	max	average	min	max
Austria	32	38	20	71	45	16	112	635	520	880
Belgium	78	41	34	47	37	32	47	692	569	815
Bulgaria	1	42			43			600		
Cyprus	11	9	2	21	6	2	16	200	59	377
Czech Republic	488	48	46	49	49	48	49	592	567	616
Denmark	83	25	8	42	21	8	34	455	280	630
Finland	42	52	23	80	42	21	62	804	622	986
France	12	51	42	61	53	37	66			
Germany	135	50	15	96	46	10	96	453	200	700
Greece	82	53	20	93	35	17	47	680	383	968
Hungary	176	56	30	148	48	33	59	556	444	815
Ireland	14	42	7	139	31	8	50	482	255	1064
Italy	192	37	3	110	30	3	97	672	160	1169
Lithuania	1	40			32			754		
Luxembourg	2	83	72	93	147	129	164	597	206	988
The Netherlands	70	38	8	74	40	8	82	532	230	1030
Poland	6	16	11	20	20	6	34	515	204	826
Portugal	10	64	37	90	52	31	72	786	473	1098
Romania	75	45	5	139	47	11	108	501	196	760
Slovakia	1	49	28	64	44	22	70	695	477	820
Slovenia	2	81	69	93	87	72	101	676	454	898
Spain	12	54	34	73	68	44	99	569	292	747
Sweden	71	75	10	98	94	7	127	734	162	960
United Kingdom	80	46	2	89	32	3	81	570	12	1000
average		47			48			598		
CV (%)		37			60			22		
overall range			2	148		2	164		12	1169

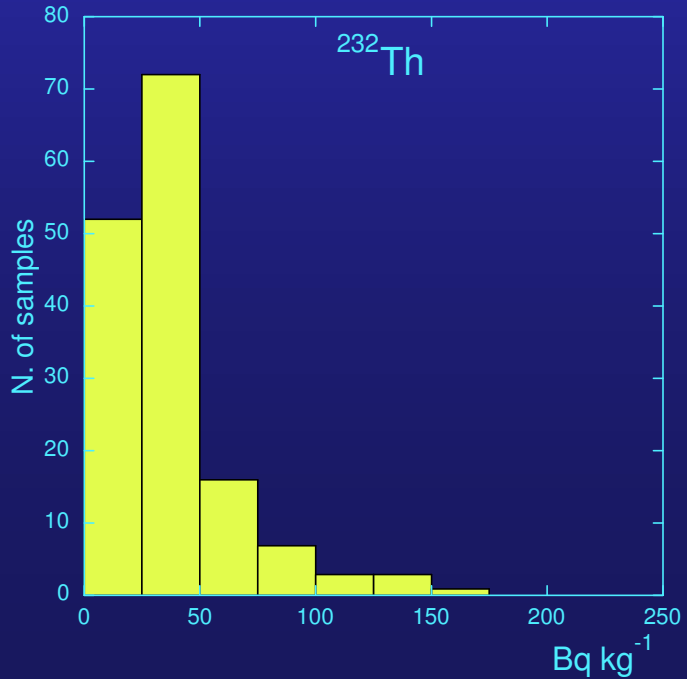
Bricks



^{226}Ra=47 ± 17



^{40}K=598 ± 132

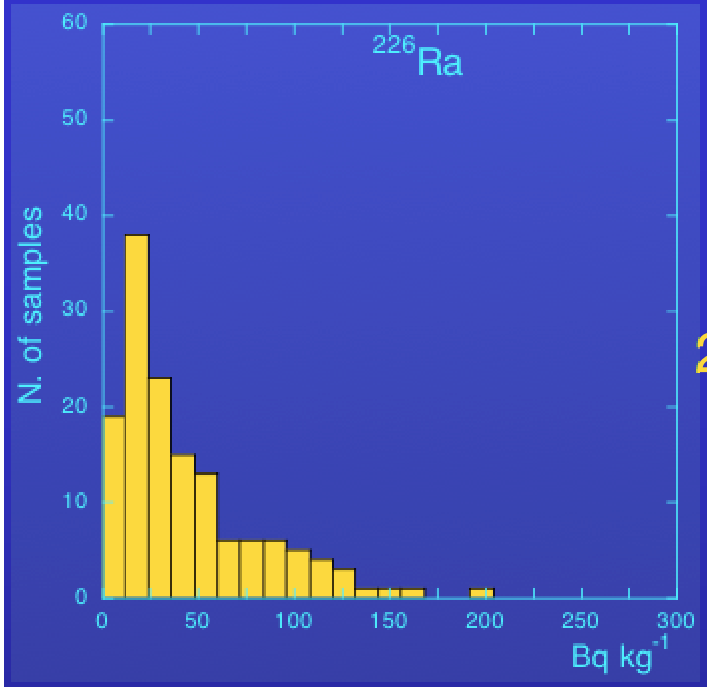


^{232}Th=48 ± 29

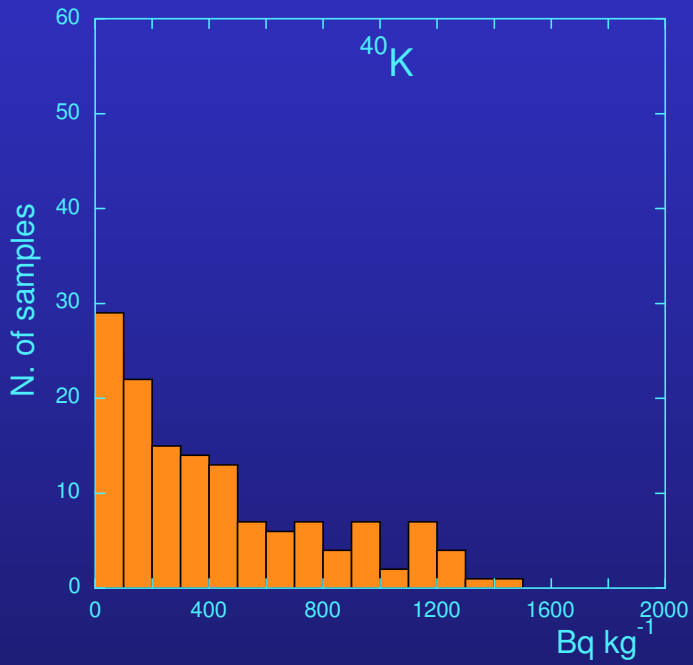
Concrete: activity concentration database (at least 2727 samples)

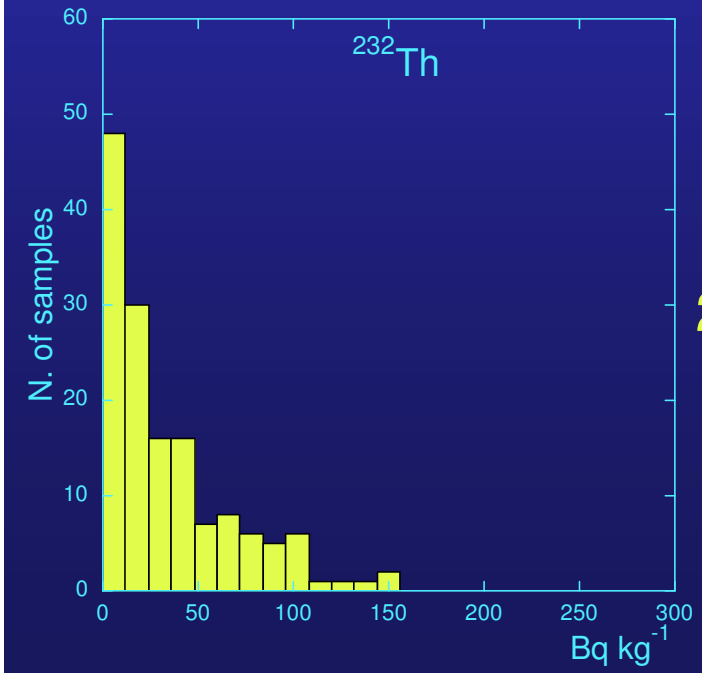
Country (23 MS)	N of samples	^{226}Ra (Bq kg^{-1})			^{232}Th (Bq kg^{-1})			^{40}K (Bq kg^{-1})		
		average	min	max	average	min	max	average	min	max
Austria	1	15	7	21	14	3	57	164	16	382
Belgium	37	17	5	42	16	5	42	247	85	490
Bulgaria	2	25	19	30	24	17	30	450	200	700
Czech Republic	491	33			24			495		
Denmark	121	152	15	670	27	10	53	620	280	1190
Finland	294	42	33	53	37	34	39	740	359	964
France	16	44	8	126	40	4	106	88	58	118
Germany	75	54	30	100	57	23	100	629	400	1100
Greece	64	40	22	85	6	3	17	101	7	383
Hungary	97	16	13	18	22	11	33	356	204	437
Ireland	8	29	18	68	12	3	13	217	16	1100
Italy	20	19	13	23	18	12	24	329	230	457
Lithuania	1	32			17			426		
Luxembourg	2	93	88	98	92	90	93	110	73	146
The Netherlands	55	35	10	115	30	6	132	263	140	870
Poland	678	115	65	200	72	36	127	666	492	1005
Portugal	38	61	1	167	50	1	152	747	11	1450
Romania	133	65	17	114	64	16	115	425	163	918
Slovakia	41	34	11	45	27	7	40	402	251	664
Slovenia	3	117	20	309	20	10	40	218	105	406
Spain	24	30			32			204		
Sweden	509	242	42	**1300**	70	31	100	627	276	819
United Kingdom	17	61	18	89	30	13	42	493	370	650
average		60			35			392		
CV (%)		90			64			53		
overall range			1	1300		1	152		7	1450

Concrete



^{226}Ra=60 ± 54





^{232}Th=35 ± 22

^{40}K=392 ± 208

Phospho-gypsum: ^{226}Ra, ^{232}Th, ^{40}K activity concentrations (at least 290 samples)

Country (11 MS)	N of samples	^{226}Ra (Bq kg^{-1})			^{232}Th (Bq kg^{-1})			^{40}K (Bq kg^{-1})		
		average	min	max	average	min	max	average	min	max
Belgium	30	431	420	442	11	10	11			
Bulgaria	2	209	18	400	17	9	25	3	1	5
Czech Republic	22	115			31			95		
Finland	17	306	24	830	23	3	118	17	9	30
Germany	2	305	60	550	20	20	20	110		
Greece	4	606	547	642	10	2	19	22	0	41
The Netherlands	20	223	28	450	24	9	48	50	16	120
Poland	28	267	61	381	17	7	28	72	41	109
Romania	73	497	155	702	40	9	89	242	44	569
Slovenia	1	500			10			41		
United Kingdom	91	1018	629	1406	33	19	48	130	41	218
overall average		407			21			78		
overall range			18	1406		2	118		0	569

^{226}Ra, ^{232}Th and ^{40}K activity concentrations in brick, concrete, cement, natural gypsum: comparison with the European soil.

Building material	Number of samples	Activity concentration (Bq kg^{-1})		
		^{226}Ra	^{232}Th	^{40}K
Brick	1676	47 (2 – 148)	48 (2 – 164)	598 (12 – 1169)
Concrete	2727	60 (1 – 1300)	35 (1 – 152)	392 (7 – 1450)
Cement	2013	45 (4 – 422)	31 (3 – 266)	216 (4 – 846)
Natural gypsum	502	15 (1 – 70)	9 (1 – 100)	91 (5 – 279)

Soil	Number of countries	Activity concentration (Bq kg^{-1})		
		^{226}Ra	^{232}Th	^{40}K
European MS average	23	36 (0 - 1000)	34 (1 – 258)	483 (0 – 3200)

corresponding to 57 (1-680) nGyh^{-1} outdoors

Distribution of bulk products exceeding the two RP 112 dose criteria

Country (24 MS)	Brick			Concrete		
	Total N of samples	N of samples exceeding dose criterion		Total N of samples	N of samples exceeding dose criterion	
		0.3 mSv y^{-1}	1mSv y^{-1}		0.3 mSv y^{-1}	1 mSv y^{-1}
		I > 0.5	I > 1		I > 0.5	I > 1
Austria	32	32	0	1	0	0
Belgium	17	17	0	29	3	0
Bulgaria	1	1	0	2	0	0
Cyprus	11	0	0	-	-	-
Czech Republic	488	488	0	491	0	0
Denmark	83	79	0	121	5	2
Finland	42	38	0	294	292	0
France	-	-	-	2	2	0
Germany	135	133	0	75	73	1
Greece	81	78	0	64	0	0
Hungary	176	176	0	97	0	0
Ireland	14	4	0	8	1	0
Italy	192	171	0	20	0	0
Lithuania	1	1	0	1	0	0
Luxembourg	2	2	2	2	2	0
The Netherlands	70	45	0	55	4	2
Poland	6	3	0	678	678	1
Portugal	10	10	0	38	27	9
Romania	75	39	1	133	110	32
Slovakia	1	1	0	41	6	0
Slovenia	2	2	1	3	1	1
Spain	12	12	0	24	0	0
Sweden	71	68	68	509	473	87
United Kingdom	71	55	7	16	6	0
total	1593	1455	79	2704	1683	135

Percentage of EU bulk products exceeding the two dose criteria of RP112

Bulk material	N. of samples with complete data set	Dose criterion (mSv y^{-1})	
		0.3	1
Brick	1593	91%	5%
Concrete	2704	62%	5%
Phosphogypsum	257	98%	84%

Percentage of bulk materials exceeding the criterion of Ö-NORM 2009

Bulk material	N. of samples with complete data set	Dose criterion
		1 (mSv y^{-1})
Brick	1593	0%
Concrete	2704	4%
Phosphogypsum	257	89%

$$I=(1+0.07\varepsilon\rho d)C_{Ra\text{-}226}/880\ Bq\ kg^{-1}+C_{Th\text{-}232}/530\ Bq\ kg^{-1}+C_{K\text{-}40}/8800\ Bq\ kg^{-1}\leq 1$$

default values: $\varepsilon = 0.1$ $\rho = 2000\ kg\ m^{-3}$ $d = 0.3\ m$

Complying with this index warrants less than 1 mSv y^{-1} due to γ and Rn from building products (natural background outdoors =1.2 mSv $^{y-1}$)

Comparison between different approaches applied to concrete

percentage of sample exceeding 1mSv y^{-1} dose criterion		
RP 112* (dose from γ)	Ö-NORM (2009)** (dose from γ & Rn)	SI 5098 (2009)** (dose from γ & Rn)
5%	4%	35%

*ρ = 2300 kg m^{-3}; d = 0.2 m

**ε = 0.1; ρ = 2300 kg m^{-3}; d = 0.2 m

Israel standard inex – SI 5098 2009

$$I=(1-\varepsilon)C_{Ra-226}/421+\varepsilon C_{Ra-226}/11.6+C_{Th-232}/298+C_{K-40}/4150 \text{ Bq} \leq 1$$

Stones as superficial materials: activity concentrationdatabase

(at least 621 samples)

Superficial stones	N of samples	^{226}Ra (Bq kg^{-1})			^{232}Th (Bq kg^{-1})			^{40}K (Bq kg^{-1})		
		average	min	max	*average*	min	max	*average*	min	max
Igneous plutonic	387	*78*	0.8	588	*89*	0.3	906	*1049*	24	2040
Igneous volcanic	86	*160*	16	709	*163*	8.0	750	*1295*	170	2354
Metamorphic	148	*27*	0.7	166	*21*	0.0	142	*395*	0.2	1891

Values of index I for superficial material

Country (15 MS)	Igneous plutonic				Igneous volcanic				Metamorphic			
	N of samples	I_{mean}	I_{max}	I_{min}	N of samples	I_{mean}	I_{max}	I_{min}	N of samples	I_{mean}	I_{max}	I_{min}
Austria	22	0.61			4	0.33			8	0.04		
Bulgaria	1	0.97							12	0.11	0.67	0.02
Cyprus	26	0.56	2.06	0.02					10	0.06	0.12	0.00
France	13								2	0.60	0.90	0.31
Germany	3	0.81	1.27	0.48	9	0.58	1.13	0.15	6	0.25	0.68	0.11
Greece	83	1.12	7.03	0.02	6	1.49	2.19	0.78	50	0.19	1.21	0.01
Italy	80	1.21	3.51	0.24	64	2.29	6.10	0.50	50	0.80	1.41	
Luxembourg	60	0.96	1.81	0.05								
The Netherlands	1	0.61			1	0.33						
Poland	1	1.10							6	0.51	0.82	0.03
Portugal	1	1.79							1	0.56		
Slovakia	1	0.72			1	0.94			1	0.11		
Spain	88	0.85	1.42	0.23					1			
Sweden	4	1.09	2.08	0.15								
United Kingdom	3	0.81	0.83	0.79								
Total	387				86				148			

Conclusions

From the analysis of this database some conclusions.

Building materials used in bulk amounts:

•Many investigations carried out in non-stony bulk products (bricks, concrete and cement) but most of the investigations were not statistically representative.

•A high variability of activity concentrations is present in bulk materials/products.

•^{232}Th activity concentration often comparable with that of ^{226}Ra, showing the need of research into measurement methods, dosimetry and the health effects of the ^{232}Th chain (Tn and Tn decay products).

•Adoption of 0.3 mSv y^{-1} dose criterion of RP112 probably too ambitious a health goal; too many materials would exceed the value of index I.

•1 mSv y^{-1} more realistic but a cost/effective goal would be a dose criterion between 0.3 and 1.

•This needs a new analysis of materials with different hypotheses and a robustness evaluation of the RP112 index I (new goal in this research activity) - BSS ”more elaborate model”.

•Data of stony material are scarce - particularly for bulk material - it is difficult to draw general conclusions.

•Some of them (e.g.tuffs) are highly radioactive and used on a local scale in some areas.

Conclusions (cont.)

By-products of industrial origin (such as fly ashes, coal ashes, slag, etc.):

- with the exception of phosphogypsum, scarse data were available due to their large heterogeneity;
- their radioactivity content is generally high, especially for ^{226}Ra;
- most of these by-products are added to cement and can cause a non-negligible increase in its radioactivity content;
- due to the limit of this database we could not present and discuss these data, or assess their radiological significance.

Superficial building materials:

- database of about 600 samples of igneous and metamorphic stones;
- only for 15 MS, not enough to give an idea of the European situation;
- data confirme
  - ✓ a high concentration of ^{226}Ra, ^{232}Th and ^{40}K in igneous stones, especially of volcanic origin
  - ✓ quite a low content of natural radionuclides in metamorphic materials, as already evidenced in bulk stony materials;
- only rarely would their use entail trade limitations.

Details of database can be found in

Trevisi R, D'Alessandro M, Risica S, Nuccetelli C J. Environ. Radioact. 105,11-20 (2012)

Thank you!