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Radiological evaluation of by-products used in construction and alternative applications; Part I. – preparation of natural radioactivity database

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Abstract

To get an insight into the radiological features of potentially reusable by-products in construction industry a review of the reported scientific data is necessary. This study is based on the continuously growing database of the By-BM (H2020-MSCA-IF-2015) project (By-products for Building Materials). Selection criteria were defined for manual data mining in such a way to avoid the collection of too heterogenous datasets. Currently, the By-BM database contains individual data of about 431 by-products and 1095 construction and raw materials. The By-BM database only consists out of measurement information on individual samples and not out of processed data that only gives a crude summary (such as only a range or average) of experimental results. As a result of the statistical analysis of the data it was found that in case of the construction materials the natural isotope content had a wider distribution than the by-products. However, the average of the Ra-226, Th-232 and K-40 contents of reported by-products were 2.00, 2.11 and 0.48, whilst the median was found 1.97, 1.24 and 0.53 times higher than the construction materials, respectively. The calculated Radium equivalent concertation was higher than the accepted value for residential properties of 370 Bq/kg in the event of 10.3% of total construction materials and 42.4% of by-

products, whilst the I-indexes were above 1.0 index value with 17.3% and 58.2%, respectively. From the obtained data, it can be concluded that generally, the reuse of industrial by-products in construction materials for residential purposes, without due diligence, can pose elevated risks to residents as a result of their high-volume usage.

Keywords: by-products; building materials; reuse; natural radiation; database; I-index; mixing

1. Introduction

The depletion of primary raw materials requires the development of new eco-innovative construction materials based on secondary resources. To counter global warming, low CO_2 emissions are a requirement to produce these new types of construction materials. The urgent investigation of reuse of by-products is essential to enable new materials to be safely and efficiently integrated into new and refurbished buildings. The revised European Union's Waste Framework Directive with its objective to reach 70% of reuse, recycling and other forms of material recovery represents the main European policy driver [1]. In October 2014 the leaders of EU (European Union) agreed on a target of 40% reduction in greenhouse gas emissions by 2030 (based on 1990 levels). This resource efficient approach is central to the circular economy. The 'end of life' concept is replaced by the concept of zero waste and the circular economy where waste production is avoided through proper design of materials, products, systems and business models resulting in many 'cascades' or cycles of use. [2]. The urgent investigation of reuse of by-products is essential to enable new materials to be safely and efficiently integrated into new and refurbished buildings. Geopolymers can be alternative low-carbon binders (produced with the reuse of industrial wastes that are produced in large quantities). The properties of geopolymers are adjustable in the function production method [3] [4]. These materials are very promising for replacing traditional construction materials and offer solution to the immobilization of toxic materials and radioactive wastes as well as the treatment of residues[5]. Construction materials can be produced directly from natural materials e.g. rocks, granite, gypsum, clay, etc. or by means of reuse of industrial by-products such as fly ash [6][7], bottom ash [8], phosphogypsum [9], steel slag [10], red mud [11], etc. Generally, the minerals contain terrestrial radionuclides from natural origin (U-238 and Th-232 series, furthermore K-40 and their progenies) which do not cause significantly higher radiation exposure than normal background levels. In soils the current worldwide average activity concentration of K-40 is 412 Bq/kg, 33 Bq/kg for U-238, 32 Bq/kg for Ra-226 and 45 Bq/kg for Th-232 [12]. In the case of the construction materials, the reported world average values are 500 Bq/kg, 50 Bq/kg, 50 Bq/kg for K-40, Ra-226 and Th-232, respectively [13]. Although the reported average activity concentrations for construction materials are relatively low, significant variation can be found from region to region. In some cases elevated level of natural radionuclides of building materials

causes significantly enhanced exposure on residents [8]. The radiation exposure originated from residential construction materials is a significant environmental factor on residents and critical receptors such as infants or the elderly that can spend 80% or even more time under indoor conditions [12]. The chronic exposure to small doses of ionizing radiation can increase the risk of health damage of people, which may occur decades after the exposure [14]. The two main exposure pathways for indoor exposure are:

1. External exposure: direct exposure of residents to gamma radiation from the naturally occurring radionuclides contained in the building materials.

2. Internal exposure: The inhaled radon (radioactive noble gas) and its progenies significantly augment the risk of the evolution of pulmonary cancer (2^{nd} risk after smoking) [15]. Radon can exhale from the soil and also from the building materials and accumulate in poorly aerated spaces, such as mines or even in buildings. Generally, the radon is the major contributor to the ionizing radiation dose received by most of the population. However, the primary source of the radon is the Ra-226 content of soil. The building materials also contribute to indoor radon depending on their Ra-226 content, porosity, and permeability.

This study is based on the continuously growing worldwide database of the By-BM (H2020-MSCA-IF-2015) project. The aim of cross-disciplinary By-BM (H2020-MSCA-IF-2015) project is to characterize the mechanical and also the radiological parameters of constituents and the prepared geopolymers (inorganic, synthetic building materials) made from industrial by-products [16] [17]. To draw conclusions from scientific data available in literature regarding the content of natural radionuclides of commercially available or newly developed construction materials and about the suitability of industrial by-products for use in building materials it is important to gather the data in a database that allow their statistical analysis and visualization. For NORM (Naturally Occurring Radioactive Materials) only a few databases exist e.g. NORM database of COST Action TU1301 NORM4Building [18] and NORM database of NIRS (National Institute of Radiological Sciences, Japan) [19]. These databases are accessible online but a drawback of these databases is that the reported information is generally, available as a range or average values of samples that are not statically related. This aspect does not enable further statistical analysis for visitors. In the case of the database constructed by Trevisi et al. of natural radioactivity in building materials in the European Union, information about more than 8000 samples were imported, evaluated and published [20].

The aims of the current study:

- Establishment of selection criteria to create a worldwide database about the natural radionuclide content of construction and raw materials and furthermore, industrial by-products

- The database only consists out of measurement information on individual samples and not out of processed data that only gives a crude summary (such as only ranges or average) of statistical unrelated experimental results.

- Statistical analysis of the reported data to obtain main statistical features (min, max, average, 1st quartile, median, 3rd quartile, distribution characteristic, box-and-whisker plot)

- Visualisation of large number of data to facilitate the comparison of different material categories

- Calculation, statistical analysis, visualisation and comparison of Radium equivalent concertation and I-indexes of imported sample information to screen materials

- To prepare the online version of By-BM database

2. Materials and methods

2.1. Restrictions set on the data that was used for the database

Generally, the reported activity concentrations of investigated samples are presented as a range with a mean value which does not allow further statistical analysis by the readers. The new database will only contain measurement information on individual samples and specific restrictions were set in order to obtain a systematic dataset suitable for statistical analysis:

- The data was imported only if it was obtained with gamma spectrometry
- Published data on individual samples was used in the database only if the Ra-226, Th-232 and K-40 contents was presented separately for each and every sample
- Average results of certain materials were used only if the investigated material originated from the same site, e.g. quarries, mines, reservoirs. In the case of commercial building materials, the brand and the type of the samples had to be clearly mentioned in the reference before the data was included. Furthermore, the range of the data was also checked and the mean was used only if the minimum and maximum values were within 20% of the mean
- In several cases, instead of the Ra-226, the U-238 activity concentration values were reported in publications. In those cases, the reported data was imported into the database only if the results were obtained from the Rn-222 progenies (Bi-214, Pb-214) to avoid the disequilibrium between U-238 and Ra-226

2.2. Classification of materials with commonly used indexes

2.2.1. Radium equivalent index

The radium equivalent index [21] (Ra_{eq}) is one of the most frequently used index calculation method to classify materials on the basis their Ra-226, Th-232 and K-40 content. Owing to the different gamma-ray emission of the terrestrial isotopes and their decay chain their dose rate contribution differs. The calculation of Ra_{eq} assumes that 259 Bq/kg of Th-232 and 4810 Bq/kg of K-40 causes an dose rate equivalent to 370 Bq/kg of Ra-226. As a result of the weighting of the dose contribution of Th-232 and K-40 isotopes, the Ra_{eq} concentration can be calculated with the following formula [21]:

$$Ra_{eq} = A_{Ra-226} + 1.43A_{Th-232} + 0.077A_{K-40} \quad (1)$$

where, $A_{Ra\text{-}226}$, $A_{Th\text{-}232}$, and $A_{K\text{-}40}$ are the activity concentration of Ra-226, Th-232, and K-40, respectively. In the case of construction materials, the Ra_{eq} concentration has to be lower than 370 Bq/kg to keep the annual external dose below 1.5 mSv/y [22]. In literature, publications are available which present differentiated application categories based on Ra_{eq} concentration ranges [8][23][24]. The Ra_{eq} concentration determines the type of allowed application. Calculation of dose rate, absorbed dose rate and annual dose excess based on RP-112.

2.2.2. Calculation of dose rate, absorbed dose rate and annual dose excess based on RP-112

A dose rate calculation method is presented in RP-112 [25] for calculating dose due to external gamma radiation from building materials. This method on the basis of the approach of Markkanen [26] is also widely used. According to the presented model described in RP-112, a reference room (dimensions of the standard room 4 m x 5 m x 2.8 m) with concrete walls (all structure such as a floor, ceiling, and walls, with 20 cm thickness and 2350 kg/m^3 density) can be used to estimate the indoor dose rate. The indoor dose rate can be obtained with the following formula:

$$\dot{D}_{indoor} = f_{Ra-226} \cdot A_{Ra-226} + f_{Th-232} \cdot A_{Th-232} + f_{K-40} \cdot A_{K-40} \quad (2)$$

Where, $A_{Ra\text{-}226}$, $A_{Th\text{-}232}$, and $A_{K\text{-}40}$ are the activity concentration [Bq/kg] of Ra-226, Th-232, and K-40, respectively. The values of conversion factors [(nGy/h)/(Bq/kg)] of Ra-226 ($f_{Ra\text{-}226}$), Th-232 ($f_{Th\text{-}232}$) and K-40 ($f_{K\text{-}40}$) are 0.92, 1.1 and 0.08, respectively.

To determine the annual dose excess [mSv/y] of residents, the natural background gamma dose rate ($\dot{D}_{background}$ = 50 nGy/h), the indoor spent time (t = 0.8 ×365 day ×24 hour, where 0.8 is the occupancy factor) and the dose conversion factor (F = 0.7 Sv/Bq) should be taken into consideration. The annual effective dose excess [mSv/y] can then be calculated with the following formula [26]:

$$E_{annual} = (\dot{D}_{indoor} - \dot{D}_{background}) \times t \times F \times 10^{-6} \quad (3)$$

2.2.3. I-index

Generally, to limit gamma exposure originated from building materials the widely used I-index – defined also in RP-112 [25] – is applied. The calculation method for the I-index is based on the model of Markkanen [26]. According to this model, an 1 mSv dose excess can be the result of an exposure to respectively 276 Bq/kg Ra-226, 231 Bq/kg Th-232 and 3176 Bq/kg of K-40. In the final formula of I-index, the values computed above are rounded to the nearest full 100 Bq/kg (Ra-226 and Th-232) or 1000 Bq/kg (K-40) [25].

$$I = \frac{C_{Ra-226}}{300 Bq/kg} + \frac{C_{Th-232}}{200 Bq/kg} + \frac{C_{K-40}}{3000 Bq/kg} \quad (4)$$

where $C_{Ra\text{-}226}$, $C_{Th\text{-}232}$, $C_{K\text{-}40}$ are the Ra-226, Th-232 and K-40 activity concentrations expressed in Bq/kg

The I-index value of 1.0 can be used as a conservative screening tool for identifying building materials that during their use would cause doses exceeding the reference level (1 mSv/y excess in addition to outdoor exposure) in the case of bulk amount inbuilt. In the European Union to control the gamma-exposure originating from building materials, the I-index is recommended for the member states to screen a defined list of materials when they are used in buildings [27]. Building products with an index larger than one the real criterion, exposure relative to the reference level (< 1 mSv/y), need to be verified and other parameters such as the density, the thickness of the material and type of their inbuilt (bulk amount or superficial) should be taken into account [28] [27].

3. Results and discussion

3.1. Database content information

The current version of the By-BM database (date: 20/12/2016) contains individual data about Ra-226, Th-232, K-40 activity concentration of 28 different materials (21 construction materials, 7 by-products, Table 1). Altogether, information about 431 by-products and 1095 construction materials and raw materials was collected from 48 countries.

The worldwide distribution and the number of data are illustrated in Fig. 1.

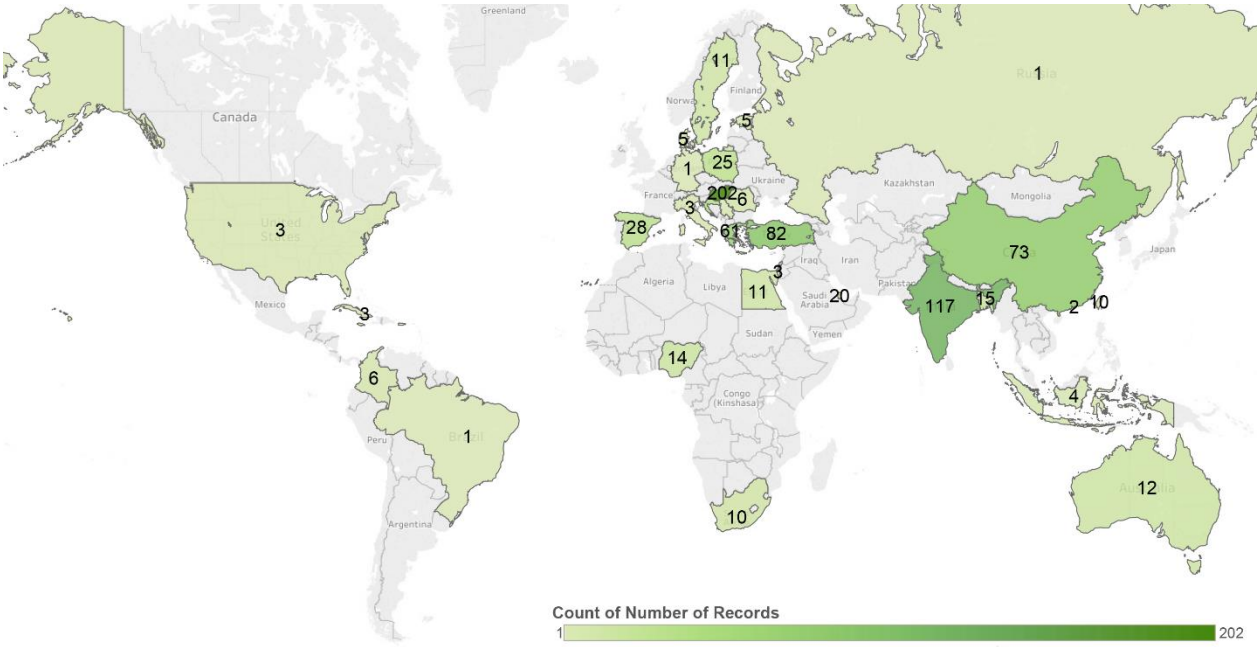


Fig. 1: The global distribution and the number of gathered data

Table 1: Number of collected materials

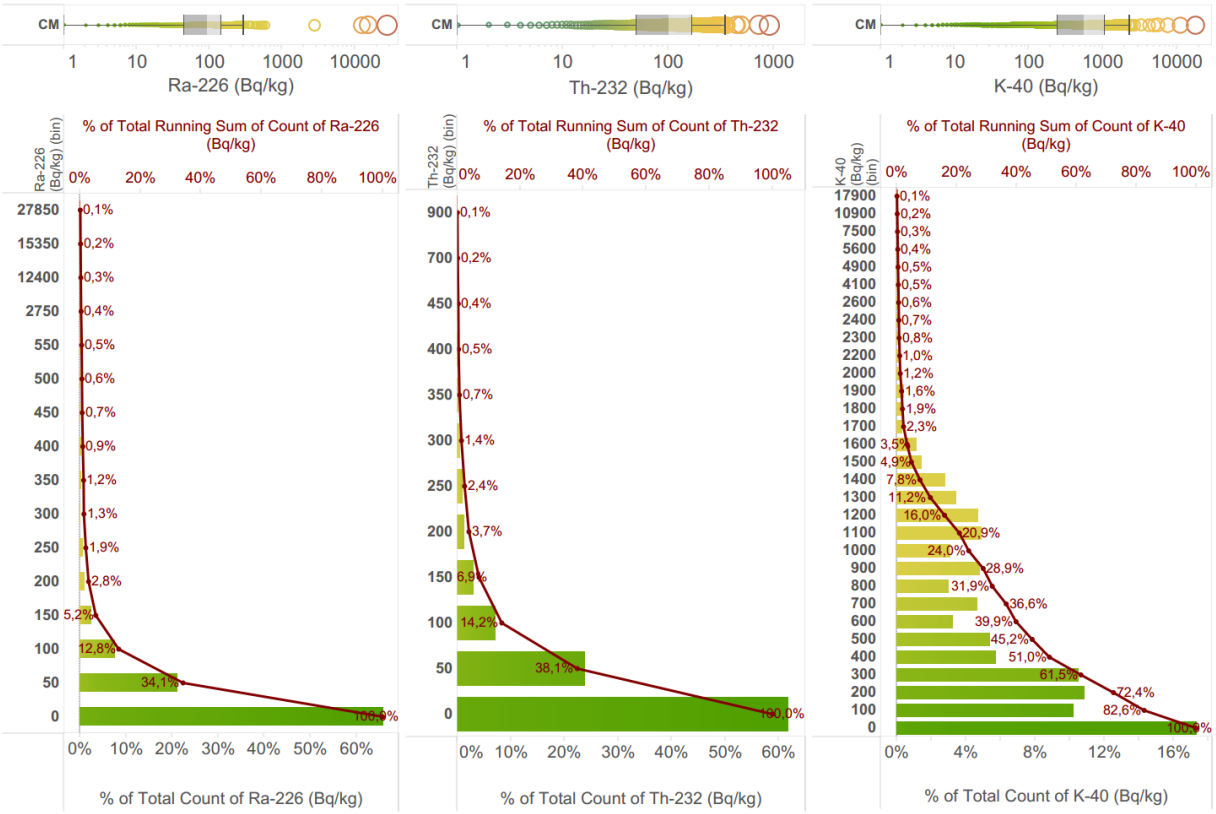
Material name	#	Reference
Aggregate	9	[29][30][31] [32]
Basalt	3	[29][33][34]
Brick	243	[35][36][37][38][39][40][29][33][24][41][42][43][44] [30][45][46][47][48][49][50][51][52][53][54][55][30][31][56]
Cement	87	[57][58][29][42][44][59] [60][48][49][50][51] [52][61][62][55][30][31][56][32]
Ceramics	94	[63][64][65][37][29][33][66][67][68][47][49][30][56]
OPC concrete	63	[35][29][33][24][41][69][70][50][71][51][30]
Gas concrete	37	[35][24][72][49][51]
Granite	297	[65][37][73][74][75][33][34][76][77][78][79][80][81][82][83][84]
Gypsum	66	[85][37][86][49][50][51][87][56][32]
Limestone	16	[29][33][34][49][50][53][31][56]
Marble	72	[37][34][33][67][47][80][50][51][83][62]
Pumice	3	[29]

Rock	31	[29][66][34][78][49][30]
Sand	19	[62][55][30][31][56][32]
Sandstone	14	[66][78][80][50]
Serizzo	5	[34]
Sienite	5	[34]
Asbestos tile	4	[55][30][31]
Travertine	9	[66][34][51]
Tuff	10	[29][66][60]
Volcanic rock	7	[83]
Bottom ash	59	[35][85][88][24][89][90][91][92][93][94]
Fly ash	145	[85][29][95][91][96][97][89][71][51][98][90][93][94][99][100]
Manganese clay	44	[101][102]
Phosphogypsum	45	[29][42][103]
Red mud	92	[104][105][58]
Steel slag	41	[106][107][108]
Residue of TiO_2	5	[66][87]

3.2. Distribution of natural radionuclide content of construction materials and by-products

The distribution of the Ra-226, Th-232, K-40 activity concentration in the event of construction materials and by-products is illustrated in Fig. 2 with 50 Bq/kg, 50 Bq/kg and 100 Bq/kg bin size resolution, respectively (empty bins were not illustrated). In the case of construction materials, the K-40 content was generally higher, while the Ra-226 and Th-232 activity levels were usually lower compared with industrial by-products.

Distribution of natural radionuclide concentration in construction materials



Distribution of natural radionuclide concentration in by-products

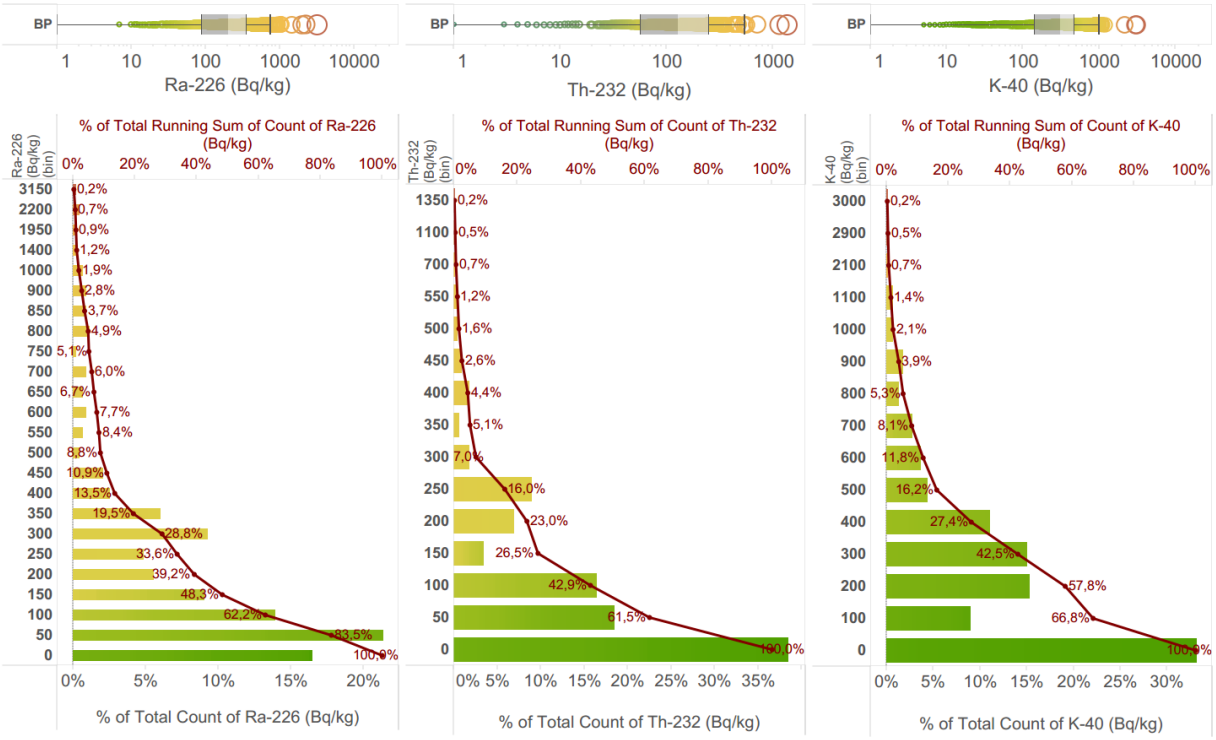


Fig. 2: Distribution of Ra-226, Th-232, and K-40 activity concentration of construction materials and by-products

As a result of the data analysis it was found that in case of the construction materials the natural isotope content varied widely, more so than for the by-products (Table 2). The obtained data was compared with world average radionuclide content of building materials (500 Bq/kg, 50 Bq/kg, 50 Bq/kg of K-40, Ra-226 and Th-232, respectively) [13]. But the average values for the Ra-226, Th-232 and K-40 content of reported by-products were respectively 2.00, 2.11 and 0.48 times higher when compared to the construction materials. The median is not skewed so much by outliers than the average values. The obtained mean values of the Ra-226, Th-232 and K-40 activity concentrations were 1.97, 1.24 and 0.53 times higher in the case of by-products. To get visual information about the distribution of the data The box-and-whisker plot diagram is widely used. In addition to the median, the lower (25 percentile) quartile, the upper (75 percentile) and 1.5×IQR (Inter Quartile Range) are also shown to detect the outliers of the dataset [109]. However, the obtained diagrams show a rough distribution of the data, further datamining could change and sophisticate the obtained picture about the natural radionuclide content in construction materials and also in by-products.

Table 2: Main statistical properties of analysed data

	Construction materials			**By-products**		
Statistical properties	Ra-226	Th-232	K-40	Ra-226	Th-232	K-40
Min [Bq/kg]	1	1	1	10	1	5
Max [Bq/kg]	27851	906	17922	3152	1350	3001
Average [Bq/kg]	112	61	668	224	129	318
Q1 Lower quartile [Bq/kg]	47	50	251	84	57	142
Median [Bq/kg]	95	101	576	187	125	307
Q3 Upper quartile [Bq/kg]	148	172	1093	338	250	471
Most frequent range [Bq/kg]	0-50	0-50	0-100	50-100	0-50	0-100
% of samples in the most frequent range	65.9	61.9	17.4	32.3	38.5	33.2
WA in building materials [Bq/kg][13]	50	50	500	50	50	500
% of samples >WA	34.1	38.1	45.3	83.5	61.5	16.2

Approximately, 34% of the collected activity concentration values for construction materials were over 50 Bq/kg Ra-226, while in the case of the by-products, 83.5% of the samples showed a Ra-226 activity concentration higher than 50 Bq/kg. The Th-232 activity concentration was over 50 Bq/kg for 38.1% of the construction materials and 61.5% of the by-products. Generally, the K-40 content in construction materials was higher than in the by-products: altogether, 45.3% of the construction materials and 16.2% of the by-

products demonstrated an activity concentration above 500 Bq/kg K-40. Although the activity concentration for K-40 is higher in the event of construction materials the formed gamma dose, as a result of their bulk amount inbuilt mainly originates from Ra-226 and Th-232 [25]. Owing to this fact, it can be stated that mainly the elevated levels of Ra-226 and Th-232 in by-products can pose an elevated risk to residents, and that therefore radiological screening of by-products is required before they are used for the production of construction or building materials.

3.3. Results of commonly used indexes

3.3.1. Radium equivalent index

In Fig. 3. and Fig. 4. detailed information is illustrated about the Ra_{eq} distribution of analysed data. It is clear from Table 3 and Fig. 3 that by-products are available that can pose elevated radiological risks when they are included in as building materials and therefore screening before use is required. Even by-products of the same plant can be quite heterogeneous [104] owing to variations in the origin of the raw materials and the applied industrial processing method. Using the classification method proposed by [8][24] [23], the population of the different categories for the Ra_{eq} activity concentrations extracted from the By-BM database is shown in Fig. 4.

The accepted Ra_{eq} ranges proposed for differentiated categories of application are the following :

I. For building residential houses: $Ra_{eq} < 370$ Bq/kg

II. For industrial use: $370 < Ra_{eq} < 740$ Bq/kg

III. For roads and railways: $740 < Ra_{eq} < 2200$ Bq/kg

IV. For landfilling: $2200 < Ra_{eq} < 3700$ Bq/kg

V. Forbidden to use for any construction: $Ra_{eq} > 3700$ Bq/kg+-

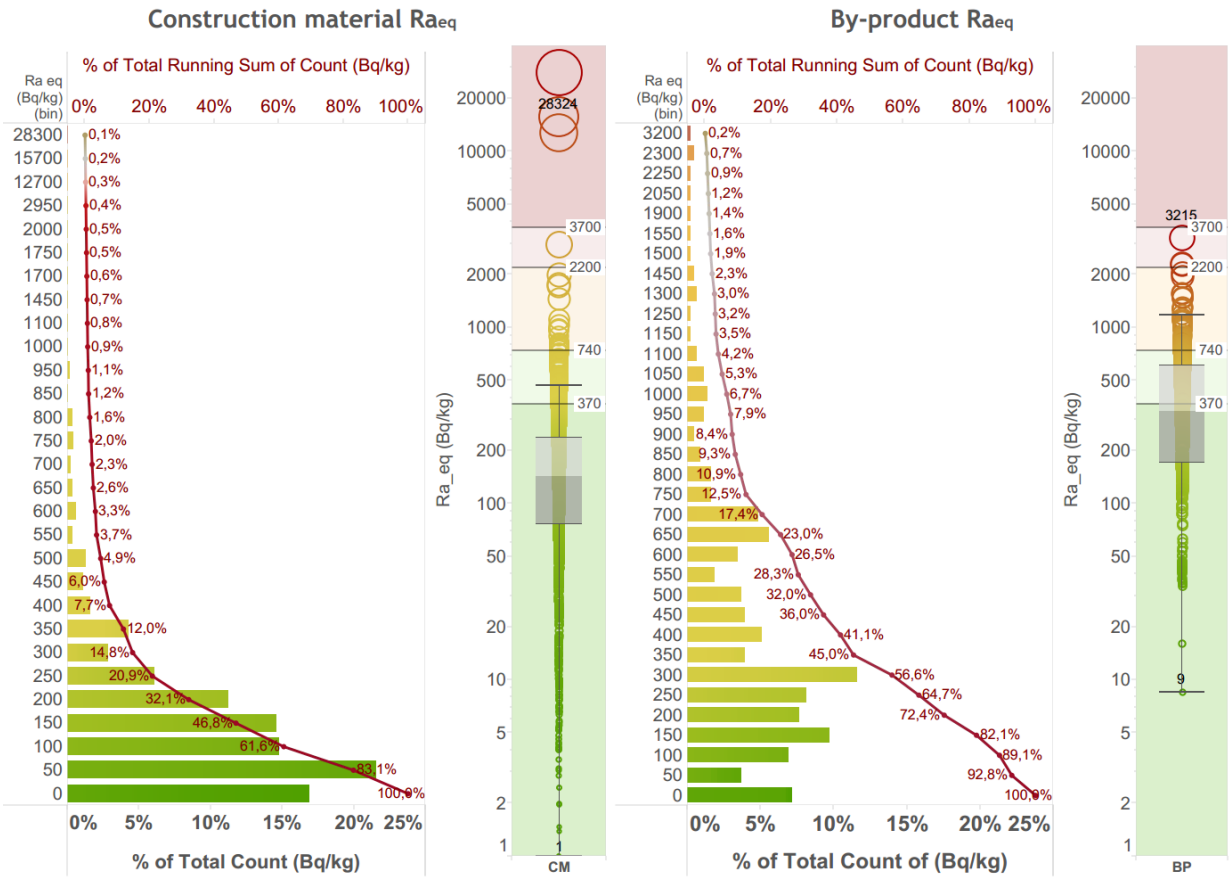


Fig. 3: Distribution of Ra_{eq} activity concentration of construction materials and by-products with 50 Bq/kg bin size resolution (empty bins were not illustrated)

The Ra_{eq} describes the gamma dose contribution of investigated materials in a more straightforward way than the plots for individual Ra-226, Th-232, K-40 activity concentrations. In Table 2. some key statistical features that were extracted from the calculated Ra_{eq} concentration values are summarised.

Table 3: Main statistical properties of the calculated Ra_{eq} concentration values

Statistical properties	**Ra_{eq} construction materials**	**Ra_{eq} by-products**
Min [Bq/kg]	1	9
Max [Bq/kg]	28324	3215
Average [Bq/kg]	233	434
Q1 Lower quartile [Bq/kg]	77	172
Median [Bq/kg]	143	334
Q3 Upper quartile [Bq/kg]	236	612
Most frequent range [Bq/kg]	50-100	300-350
% of samples in the most frequent range	21.5	11.6
% of samples > 370 Bq/kg Ra_{eq}	10.3	42.4

For construction materials, 89.7% can be found in category I. under 370 Bq/kg Ra_{eq}. In the higher categories, a limited amount of granite and rock samples were found while for the other considered construction materials over 95% of the construction materials were found under category I. In total, only 28.3% of the granite and only 11.4% of the rock were found under categories II, III, IV and V. On the contrary, numerous by-product samples – except manganese clay – needed to be categorised in category II, III or IV.

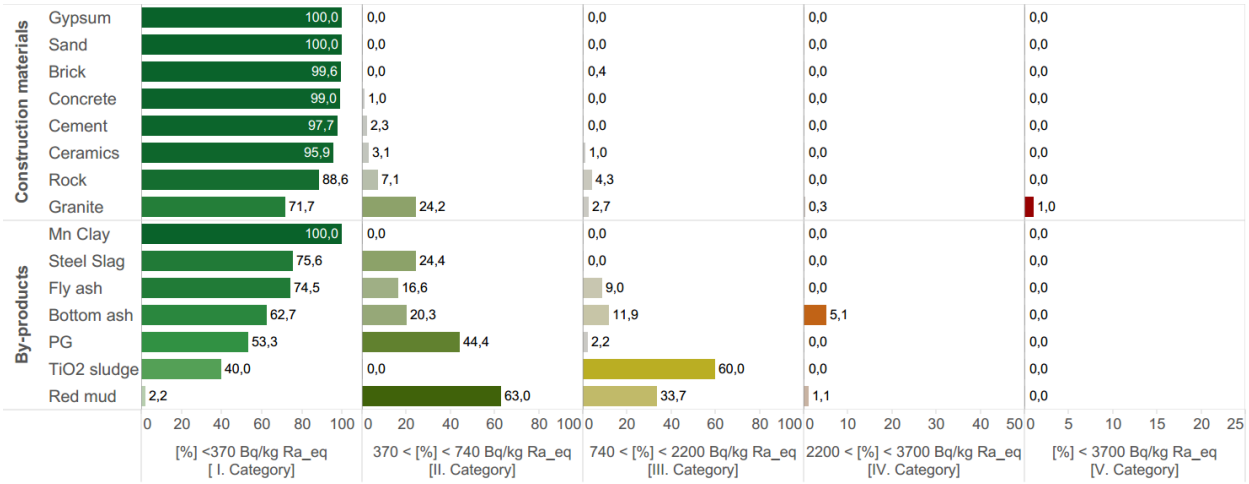


Fig. 4: Distribution of Ra_{eq} activity concentration of construction materials and by-products according to categorized classification criteria

For red mud the main fraction, 63% was found under category II, dedicated to industrial construction, while for phosphogypsum this was 44.4% and for steel slag 24.4%. In category III, between 740 and 2200 Bq/kg Ra_{eq}, a significant amount of records related to by-products was found, especially TiO_2 sludge (60%), red mud (33.7%), bottom ash (11.9%) and fly ash (9%), which would be eligible only for road construction. Only a very limited amount of data (5.1% of bottom ash and 1.1% of red mud) was found between 2200 and 3700 Bq/kg Ra_{eq} (category IV) which indicates that the materials are still acceptable for landfilling. Above 3700 Bq/kg Ra_{eq}, only 3 Egyptian granites (1.0% of total granite samples) were found, which cannot be used for any kind of construction applications.

3.3.2. Annual effective dose excess based on RP-112

The annual effective dose excess based on the dose calculation method presented in RP-112 [25] is shown in Fig. 5. It can be clearly seen that in the case of construction materials more than 84% of the reported data was lower than the reference level with 1.0 mSv annual dose excess, whilst in the case of by-products only 41% was in a lower dose excess assuming bulk incorporation. It was also found that 25% of the construction materials could cause less than 50 nGy/h absorbed gamma dose rate (AGDR, Fig. 5). It means that the bulk incorporation would result in a lower absorbed gamma dose rate relative to the world average background radiation.

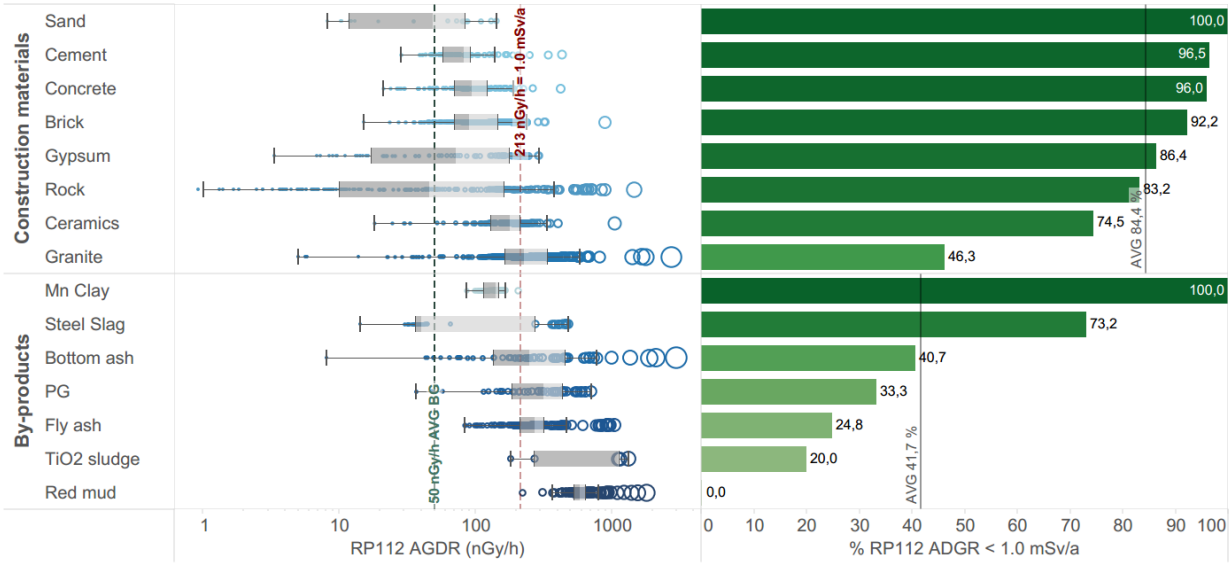


Fig. 5: Absorbed gamma dose rate and annual gamma dose excess of analyzed construction materials and by-products based on RP-112 [25]

3.3.3. I-index

Regarding to the I-index it should be noted that the I-index can be used only for real building materials such as concrete and ceramics (but not for cement). The calculation of I-index of any raw (sand, aggregate), construction materials (cement, lime) or by-products would imply that 100% of these materials are used as building materials. Of course, this not realistic for most construction materials where only a fraction of a certain by-products can be included, but it provides opportunity for their screening or prediction [6] of the I-index of the final product. If needed a dilution factor could be used for a given application to achieve more realistic screening (Fig.7). The main parameters obtained after the statistical analysis of the calculated I-index values, related to construction materials and by-products, are presented in Table 4.

Table 4: Main statistical properties of calculated I-indexes

Statistical properties	**I-index of construction materials**	**I-index of by-products**
Min	0.01	0.03
Max	94.6	10.7
Average	0.84	1.50
Q1 Lower quartile	0.29	0.63
Median	0.56	1.18
Q3 Upper quartile	0.89	2.11
% of samples > 1.0 I-index	17.3	58.2

The calculated I-indexes (for the by-products the assumption was made that 100% of the by-products used as a building material) are shown in Fig. 6. As it was expected from the radionuclide distribution and from calculated Ra_{eq} in the event of the by-product the calculated I-index values were significantly higher than construction materials. In Fig. 6 it can be clearly seen that all the by-products, except the manganese clay, can give yield to an I-index higher than 1.0 in numerous cases.

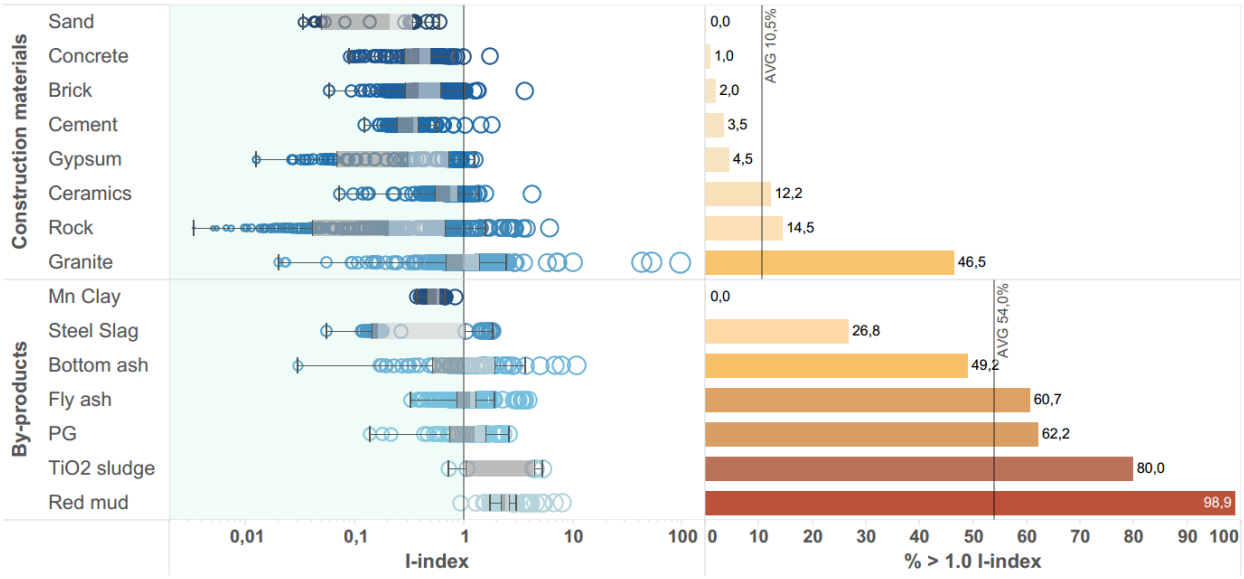


Fig. 6: I-index of analyzed construction materials and by-products

The red mud samples had a higher value than 1.0 for almost all entries, the TiO_2 sludge was also higher in the case of 80% of entries.

3.3.4. Estimation of maximum allowable mixing ratio of by-products in building materials based on I-index

To estimate an allowable mixing ratio for industrial by-products a simple mixing calculation was performed. The calculation was based on the assumption that the other components of the mixture an activity concentration that coresponds to the world average activity concentration of building materials (500 Bq/kg, 50 Bq/kg, 50 Bq/kg for K-40, Ra-226 and Th-232, respectively) presented in UNSCEAR 1993 [13]. For the mixing calculation the following equation was used:

$$X_{BP\%} \times I_{BP} + X_{WA\%} \times I_{WA} = X_{100\%} \times I_{\max_1.0} \quad (5)$$

where $X_{BP\%}$; $X_{WA\%}$ and $X_{100\%}$ are the ratio in % of the by-products, other components (aggregactes, bindres, etc) of the mixture with world average radionuclide activity concentration (hypotethic value) and the total amount (100%), respectively. The I_{BP}, I_{WA} are the calculated I-indexes of by-products and the world average radionuclide content. The $I_{max_1.0}$ is 1.0 I-index value used as upper level reference for screening. The obtained maximal allowable mixing ratios are illustrated in Fig. 7. Of course the by-

products with I-index value ≤ 1.0 can be mixed without any restriction but above that value the maximum allowable mixing ratio should be calculated.

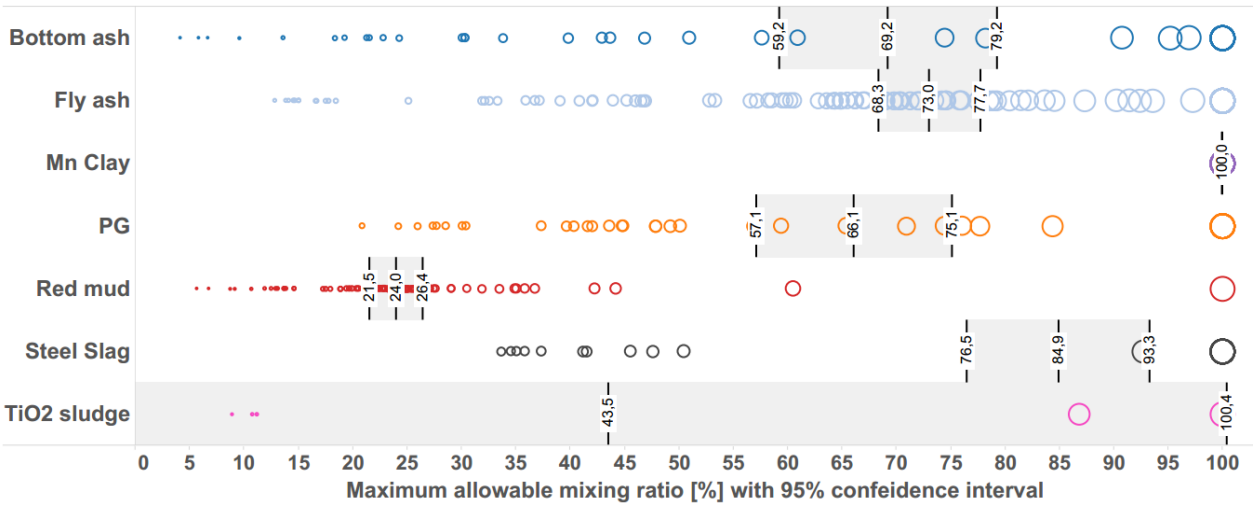


Fig. 7: Maximum allowable mixing ratio of by-products related to world average radionuclide content of building materials [13]

To draw conlusions regarding the inhomogeinity of natural radiouclide content of by-products and also about their mixability the weighted average values were also computed with 95% confidence interval. The width of the intervals depends on the number of the data analyses and also on the inhomogeneity of the samples but this parameter can be used to facilitate conclusions about the inhomogeneity. The narrowest interval (±2.5% width) was found in the case of red mud around its 24.0% average mixing ratio. Also a narrow range, only ±4.7% around its 73.3% average was observed related to fly ash. However, the confidence intervals combined with the inhomogeneity of some materials can be roughly estimated to make overall conclusion according to the number of analysed data. As the database increases a more clear picture will develop about the worldwide distribution of their radionuclide content. According to I-index values of by-products, it can be concluded that generally, the reuse of them as building materials is possible, but only in limited amounts with regular screening. The radionuclide content of recycled by-products cannot be ignored since they can cause increased radiological risk. According to EU-BSS [27], the dilution factor has to be determined as the function of the activity concentration of the components [6] [104].

The screening of by-products and construction materials can be a practical tool to identify and manage potential material resources which can pose elevated risk. The bulk amount of these materials included in the design of building products requires more specified design [58] with e.g. density and thickness characterization as highlighted in EU-BSS [27]

4. Conclusion

A large disparate quantity of data regarding the natural radioactivity is reported in literature. Generally, this data is presented as ranges or in diagrams which makes it less suitable for detailed further statistical analysis. However, there is also data available involving individually reported sample information and this data was processed for statistical analysis.

As a result of the statistical analysis it was found that the Ra-226, Th-232 and K-40 content of reported recycled by-products was 2.00, 2.11 and 0.48 times higher compared with natural construction materials. It can be concluded that some of the studied by-products can pose elevated radiological risks in cases where they are included as building materials, and therefore screening before their reuse is required. The calculated Ra_{eq} and I-indexes are useful tools to classify materials before inclusion in building products. However, these indexes can exemplify the risk of the external exposure better than the activity concentration of Ra-226, Th-232, K-40. Other factors (density and thickness) should be taken into consideration when designing building materials that contain such recycled by-products. Furthermore, differentiated categories could offer more flexible reuse options depending on the final use. However, the statistical results of current study provide the possibility to make the first rough conclusions about the worldwide radionuclide content of construction materials and industrial by-products, as the database increases this will lead to a clearer picture of the distribution of radionuclides in surveyed materials, which can be obtained with further data mining based on the established unified selection criteria.

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