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

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HIGHLIGHTS

- Manual datamining was used to gather separately reported sample information.
- 431 by-products and 1095 construction materials were collected from 48 countries.
- Ra-226, Th-232 content were 2.00 and 2.11 times higher in the case of by-products.
- Ra equivalent concentration was 1.86 times higher in the case of by-products.
- I-index >1.0 in the event of 17% of construction materials; 58% of by-products.

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ABSTRACT

To get an insight into the radiological features of potentially reusable by-products in the 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 heterogeneous 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 rough summary (such as only a range or average) of experimental results. As a consequence of the statistical analysis of the data, it was found that in the 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, while the median was found 1.97, 1.24 and 0.53 times higher than the construction materials, respectively. The calculated Radium equivalent concertation was greater 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, while 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 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.

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 to 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.

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The "end of pipe" 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 a solution to the immobilisation 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. 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 small, significant variation can be found from region to region. In some cases, an 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 ionising radiation can increase the risk of health damage of people, which may occur decades after the exposure [14]. The two most important 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 (2nd 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. The radon is the major contributor to the ionising 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 characterise 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 the 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 visualisation. 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 statistically 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 was imported, evaluated and published [20].

The aims of the current study:

- Establishment of selection criteria to create a worldwide database of 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 rough 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 to obtain a systematic dataset suitable for statistical analysis:

- The data was imported only if it was obtained by 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 explicitly 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 methods 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 a 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-226}, A_{Th-232}, and A_{K-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.