

Natural radioactivity in cements and raw materials used in Albanian cement industry

Ferat Shala¹ · Gerti Xhixha² · Merita Kaçeli Xhixha³ · Fadil Hasani⁴ · Elona Xhixha⁵ · Manjola Shyti⁶ · Dhurata Sadiraj Kuqi⁷ · Dritan Prifti⁶ · Mevlan Qafleshi⁷

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Abstract This study examines the natural radioactivity in cements and raw materials used in cement production industry with the aim to assess the population exposure to the radiation due to their use in dwellings. The results show that the limestone is the principal contributor for the presence of ²²⁶Ra activity concentration in cements. The contribution of aluminosilicate materials, mainly for the presence of ⁴⁰K and ²³²Th, shows high variability mainly due to the possibility of use of different types of raw materials. The activity concentration is found to be higher in Portland cements relative to Portland-limestone cements with values varying from 12.0 to 16.1 Bq kg⁻¹ for ²²⁶Ra, 46.2–51.2 Bq kg⁻¹ for ²³²Th and 133.7–168.8 Bq kg⁻¹ for ⁴⁰K. The activity concentration index (ACI), recommended by the Council Directive 2013/59/EURATOM and recently

amended in the Albanian legislation, was adopted as a screening tool to assess the radiological hazard due to the use of cements as building materials. The ACI varies from 0.26 to 0.31, which allows us to conclude that potentially these building materials do not pose any significant risk to humans due to their use in dwellings. This conclusion is confirmed by calculating the annual effective dose rate, varying from 0.33 to 0.38 mSv y⁻¹.

Keywords Natural radioactivity · Portland cement · Gamma spectrometry · Radiation exposure · Effective dose equivalent

Introduction

Cement production industry plays an important role in the national economy being a widely used constituent material for the production of concrete, plaster and concrete blocks. Cement is a composite building material derived from rocks and industrial by-products; therefore, it may contain varying amounts of trace elements and among them natural radionuclides of ²³⁸U and ²³²Th (and their decay progeny), and ⁴⁰K. The concentration of natural radionuclides in raw materials can vary considerably according to the geological locations and geochemical characteristics of the materials (Yuce et al. 2009; Callegari et al. 2013; Strati et al. 2014; Kaçeli Xhixha et al. 2016). Therefore, the determination of the natural radioactivity level in raw materials is important for understanding the presence of radionuclides in building materials in order to have control on the production of cements having relatively high radionuclide concentration as reported in Baykara et al. (2011).

Moreover, the knowledge of natural radioactivity in building materials is important for the determination of the

✉ Gerti Xhixha
xhixha@fe.infn.it

¹ Faculty of Mechanical Engineering, University of Pristina “Hasan Prishtina”, Bregu i Diellit, 10000 Pristina, Kosovo

² Department of Physics, Faculty of Natural Sciences, University of Tirana, Blvd. Zogu I, 1001 Tirana, Albania

³ Department of Engineering, Faculty of Professional Studies, University “Aleksandër Moisiu” Durrës, Str. Currila 1, 2000 Durrës, Albania

⁴ Kosovo Agency for Radiation Protection and Nuclear Safety (KARPNS) - Office of the Prime Minister, Ish-Gërmia, 10000 Pristina, Kosovo

⁵ Center for GeoTechnologies, University of Siena, Via Vetri Vecchi 34, 52027 San Giovanni Valdarno, Arezzo, Italy

⁶ Institute of Applied Nuclear Physics, University of Tirana, Rr. Thoma Filipeu, P.O. Box 85, Qesarakë, Tirana, Albania

⁷ Faculty of Engineering Mathematics and Engineering Physics, Polytechnic University of Tirana, Square Nënë Tereza, 4, Tirana, Albania

indoor population exposure to radiation since people spend about 80% of their time in dwellings (UNSCEAR 2000). The recent basic safety standard laid down by the Council Directive 2013/59/EURATOM (2014) requires a screening control of building materials by applying a reference level for indoor external exposure to gamma radiation emitted by building materials of 1 mSv y^{-1} , in addition to outdoor external exposure. The screening consists in the calculation of an activity concentration index (ACI), based on the determination of the activity concentration of ^{226}Ra , ^{232}Th and ^{40}K in building materials.

Previous study on the natural radioactivity in building materials does not consider the different type of cements produced in Albania (Xhixha et al. 2013a). Moreover, there is a lack of knowledge regarding the presence of radionuclides in different cement types and the relative contribution of raw materials used. Therefore, the main objective of this survey is to examine the raw materials in order to study their relative contributions to the natural radioactivity concentration in different cement types. These results are used to assess the potential health implications due to the use of such cement products in constructions.

Materials and methods

Sample collection and preparation

The cement production industry requires a close control of the chemistry of raw materials that is essential for producing different types of cement. The typical raw materials used for clinker production are sampled directly from the industrial streams: In particular, 20 samples of limestone, flysch, bauxite and pyrite are collected in order to investigate the presence of natural radionuclides. The chemical composition of clinker that exits the rotary kiln is strictly standardized in order to have similar properties with that produced by different manufacturers. The clinker is then ground with limestone and gypsum in different fractions, to produce different cement types, which are standardized according to EN 197-1 (2002). The cement type CEM I, well known as Portland cement, is produced by grinding clinker (95–100%) and minor constituents, e.g., natural gypsum (5–0%). Two types of Portland-limestone cements CEM II/A-LL and CEM II/B-LL are also produced, containing 6–20% and 21–35% limestone, respectively, with additional minor constituents (EN 197-1 2002). The samples of clinker, gypsum and three types of cement are collected (25 samples) immediately after production.

Samples are homogenized (except for cements) to a particle size of less than 2 mm and then are dried for at least 24 h at a temperature of $105 \text{ }^\circ\text{C}$ until achieving a

constant weight. Then samples are transferred into cylindrical polycarbonate boxes with a volume of 180 cm^3 and are sealed hermetically. Sealed samples are left undisturbed for at least four weeks, prior measuring them by using high-purity germanium (HPGe) gamma-ray spectroscopy, in order to establish a radioactive equilibrium in the ^{226}Ra decay chain segment.

Gamma-ray spectrometry measurements

Samples are measured using high-resolution gamma-ray spectrometry consisting of two 60% relative efficiency coaxial p-type HPGe gamma-ray detectors, with an energy resolution of approximately 1.9 keV at 1332.5 keV (^{60}Co) (Xhixha et al. 2013b). The absolute full energy peak efficiency of the MCA_Rad is calibrated using certified reference materials of natural origin. The overall uncertainty in the efficiency calibration is estimated to be less than 5% (Xhixha et al. 2016a).

The ^{226}Ra activity concentration is determined through the two main gamma emissions of radon progenies of ^{214}Pb (at 352 keV) and ^{214}Bi (at 609 keV). The ^{232}Th activity concentration is determined by assuming secular equilibrium with ^{228}Ra and ^{228}Th : The ^{228}Ra is determined through its direct progeny, ^{228}Ac gamma emissions (at 338 and 911 keV), while ^{228}Th activity concentration is determined by analyzing the two main gamma emissions of radon progenies of ^{212}Pb (at 239 keV) and ^{208}Tl (at 583 keV). Instead, the activity concentration of ^{40}K is determined from its only gamma emission at 1460 keV. The participation in the intercomparison exercises organized in the frame of COST Action-TU1301 “NORM4Building” is used for quality control purposes (Xhixha et al. 2016b).

Results and discussion

Activity concentrations

In Table 1 are shown the range and the average activity concentrations of ^{40}K , ^{226}Ra and ^{232}Th with their standard deviation (for a confidence level of $k = 1$) for the raw materials, clinker and different cement types. The activity concentration of ^{40}K , ^{226}Ra and ^{232}Th in raw materials shows a high variability. The activity concentration of ^{226}Ra in bauxite is found to be relatively high ($78.7 \pm 4.4 \text{ Bq kg}^{-1}$), approximately twice that found in limestone ($38.0 \pm 8.5 \text{ Bq kg}^{-1}$); however, considering that bauxite is used in relatively small amounts, it is reasonable to assume that limestone gives the principal contribution to the activity concentration of clinker and cement. On the other hand, flysch is expected to give the

Table 1 Average activity concentration together with their ($k = 1$) standard deviation and the range of values (in parenthesis) for ^{40}K , ^{226}Ra and ^{232}Th in raw materials, clinker and different cement types

Material type	^{40}K (Bq kg $^{-1}$)	^{226}Ra (Bq kg $^{-1}$)	^{232}Th (Bq kg $^{-1}$)
Raw materials			
Limestone	6.5 ± 5.2 (2.6–13.5)	38.0 ± 8.5 (26.4–45.9)	3.3 ± 1.4 (2.3–5.0)
Flysch	485.2 ± 126.6 (342.2–595.2)	21.2 ± 3.2 (17.0–24.1)	33.4 ± 6.8 (25.7–40.1)
Bauxite	234.8 ± 17.8 (226.4–259.1)	78.7 ± 4.4 (75.8–84.6)	108.0 ± 5.6 (103.2–112.8)
Pyrite	139.8 ± 11.2 (130.4–153.9)	6.7 ± 1.5 (5.1–8.6)	3.1 ± 1.0 (2.3–3.7)
Intermediate material			
Clinker	160.3 ± 26.4 (140.0–196.4)	55.5 ± 2.5 (53.1–57.3)	17.0 ± 2.8 (13.7–20.0)
Composite materials			
Gypsum	66.8 ± 11.3 (53.7–79.8)	11.8 ± 2.0 (9.7–14.3)	5.8 ± 1.9 (3.3–7.0)
Limestone*	6.5 ± 5.2 (2.6–13.5)	38.0 ± 8.5 (26.4–45.9)	3.3 ± 1.4 (2.3–5.0)
Cement types			
CEM I (42.5R)	168.8 ± 24.6 (140.0–185.7)	51.2 ± 5.5 (46.1–57.6)	16.1 ± 2.3 (14.0–18.1)
CEM II/A-LL (42.5 R)	150.4 ± 19.8 (119.8–165.8)	51.0 ± 3.7 (48.0–55.4)	16.5 ± 3.6 (13.5–21.5)
CEM II/B-LL (32.5R)	133.7 ± 11.4 (121.6–143.6)	46.2 ± 3.6 (41.5–49.0)	12.0 ± 3.1 (9.4–15.0)

* The same with limestone as raw material

main contribution to the activity concentration of ^{40}K and ^{232}Th of clinker and consequently cement. The activity concentrations of ^{226}Ra and ^{232}Th in limestone are comparable with other studies, while the activity concentration of ^{40}K is lower in comparison with most of the cases (Table 2). A low variability is observed also for gypsum samples, where the reported activity concentrations are generally comparable. Instead, the silica, alumina and iron-bearing raw materials show high variability of activity concentrations of ^{40}K , ^{226}Ra and ^{232}Th mainly because of different lithology and geological area.

The clinker and cement (focusing similar types of cements) show relatively variable activity concentrations (Table 2), generally comparable for ^{40}K and ^{232}Th , while the activity concentrations of ^{226}Ra are relatively higher. The average activity concentration in clinker is found to be $160.3 \pm 26.4 \text{ Bq kg}^{-1}$ for ^{40}K , $55.5 \pm 2.5 \text{ Bq kg}^{-1}$ for ^{226}Ra and $17.0 \pm 2.8 \text{ Bq kg}^{-1}$ for ^{232}Th (Table 1). The activity concentration of ^{40}K in different cement types is found to be from $133.7 \pm 11.4 \text{ Bq kg}^{-1}$ (CEM II/B-LL) to $168.8 \pm 24.6 \text{ Bq kg}^{-1}$ (CEM I). The same trend is observed for ^{226}Ra and ^{232}Th , where their activity concentrations are found to be relatively higher in CEM I and lower in CEM II/B-LL, respectively, from 51.2 ± 5.5 to $46.2 \pm 3.6 \text{ Bq kg}^{-1}$ for ^{226}Ra and from $16.1 \pm 2.3 \text{ Bq kg}^{-1}$ to $12.0 \pm 3.1 \text{ Bq kg}^{-1}$ for ^{232}Th . These evidences give a clue about the relationship between the activity concentrations of ^{40}K , ^{226}Ra and ^{232}Th in cement and the activity concentration in clinker and additive materials.

In order to quantify the contribution of raw materials, we focused on the limestone as primary constituent

material and investigated the ratio between the activity concentrations between cement and limestone in different available studies. Applying robust statistical method, we obtain 1.4 ± 0.2 for ^{226}Ra and 2.3 ± 1.5 and 2.3 ± 1.6 , respectively, for ^{232}Th and ^{40}K (Fig. 1), by calculating the median and robust standard deviation [RSD = $1.5 \cdot \text{MAD}$ (median absolute deviation)] (ISO/IEC 13528 2005). These results indicate a good correlation for the activity concentration of ^{226}Ra in cement and limestone. Furthermore, the ratio value close to unity shows that limestone is the principal contributor, approximately 70%, for the presence of ^{226}Ra in cement. On the other hand, the respective ratios for ^{232}Th and ^{40}K in cement and limestone show that aluminosilicate raw materials are instead the principal contributors for the presence of ^{232}Th and ^{40}K in cement. These materials are characterized by a higher variability because of the different geological areas and availability of different types of raw materials.

Assessment of radiological hazard

According to the Council Directive 2013/59/EURATOM (2014), the reference level applying to indoor external exposure to gamma radiation emitted by building materials, in addition to outdoor external exposure, shall not exceed 1 mSv y^{-1} . The radiological hazards due to building materials can be conservatively investigated by assessing the activity concentration index (ACI) according to the following equation:

$$\text{ACI} = A_{226\text{Ra}}/300 + A_{232\text{Th}}/200 + A_{40\text{K}}/3000 \leq 1 \quad (1)$$

Table 2 Comparison of mean activity concentrations of ^{40}K , ^{226}Ra and ^{232}Th in Albanian cement types and raw materials with available results reported in other countries

Author	Country	Material type	^{40}K (Bq kg $^{-1}$)	^{226}Ra (Bq kg $^{-1}$)	^{232}Th (Bq kg $^{-1}$)
This study	Albania	Limestone	6.5 ± 5.2	38.0 ± 8.5	3.3 ± 1.4
Tufan and Disci (2013)	Turkey	Lime	88 ± 7	20 ± 5	20 ± 9
Turhan (2008)	Turkey	Limestone	88.1 ± 29.1	16.5 ± 12.5	7.7 ± 7.2
Khan and Khan (2001)	Pakistan	Limestone	73.8 ± 21.9	21.9 ± 7.6	8.6 ± 1.6
Papaefthymiou and Gouseti (2008)	Greece	Limestone	101 ± 9	6.0 ± 0.4	6.6 ± 1.0
El-Taher et al. (2010)	Egypt	Limestone	61.2 ± 3.1	19.7 ± 2.9	39.0 ± 2.0
Aslam et al. (2012)	Pakistan	Limestone	63.1 ± 17.3	28.4 ± 8.7	11.3 ± 1.7
Al-Dadi et al. (2014)	Saudi Arabia	Limestone	< MDA	42.8 ± 0.4	0.9 ± 0.1
This study	Albania	Flysch	485.2 ± 126.6	21.2 ± 3.2	33.4 ± 6.8
Turhan (2008)	Turkey	Clay	629.3 ± 232.1	26.7 ± 20.0	41.8 ± 27.7
	Turkey	Marl	316.3 ± 131.6	17.1 ± 7.7	14.9 ± 7.1
Khan and Khan (2001)	Pakistan	Slate	821.5 ± 86.0	25.6 ± 7.7	43.2 ± 3.9
		Laterite	311.0 ± 55.5	104.6 ± 7.6	129.9 ± 7.9
El-Taher et al. (2010)	Egypt	Clay	130.7 ± 6.5	33.7 ± 4.4	68.9 ± 3.5
Aslam et al. (2012)	Pakistan	Clay	187.6 ± 17.2	34.7 ± 13.1	41.2 ± 6.7
		Laterite	195.4 ± 29.2	41.1 ± 11.8	39.3 ± 6.9
Al-Dadi et al. (2014)	Saudi Arabia	Clay	127 ± 1.8	18.2 ± 0.2	22.4 ± 0.3
		Sandstone	56.4 ± 1.6	6.2 ± 0.2	4.7 ± 0.2
This study	Albania	Bauxite	234.8 ± 17.8	78.7 ± 4.4	108.0 ± 5.6
Turhan (2008)	Turkey	Bauxite	43.2	17.1 ± 2.4	19.8 ± 0.9
This study	Albania	Pyrite	139.8 ± 11.2	6.7 ± 1.5	3.1 ± 1.0
Turhan (2008)	Turkey	Pyrite	35.2 ± 20.8	8.8 ± 9.5	5.9 ± 6.9
El-Taher et al. (2010)	Egypt	Iron oxide	121.3 ± 6.1	160.5 ± 10.5	87.3 ± 4.5
Al-Dadi et al. (2014)	Saudi Arabia	Iron	44.8 ± 1.4	37.2 ± 0.3	28.8 ± 0.4
This study	Albania	Clinker	160.3 ± 26.4	55.5 ± 2.5	17.0 ± 2.8
Turhan (2008)	Turkey	Clinker	219.0 ± 45.2	28.3 ± 13.3	15.9 ± 3.2
Papaefthymiou and Gouseti (2008)	Greece	Clinker	141 ± 13	15 ± 1	14 ± 3
Aslam et al. (2012)	Pakistan	Clinker	258.4 ± 15.3	51.1 ± 18.2	23.2 ± 1.2
Al-Dadi et al. (2014)	Saudi Arabia	Clinker	6.1 ± 0.4	79.9 ± 0.5	7.5 ± 0.3
This study	Albania	Gypsum	66.8 ± 11.3	11.8 ± 2.0	5.8 ± 1.9
Tufan and Disci (2013)	Turkey	Gypsum	35 ± 5	8 ± 3	11 ± 7
Turhan (2008)	Turkey	Gypsum	44.5 ± 23.2	10.8 ± 12.2	3.6 ± 2.7
Khan and Khan (2001)	Pakistan	Gypsum	173.7 ± 50.3	6.2 ± 1.6	13.3 ± 2.8
Papaefthymiou and Gouseti (2008)	Greece	Gypsum	< MDA	6.8 ± 0.6	< MDA
El-Taher et al. (2010)	Egypt	Gypsum	88.7 ± 4.4	31.7 ± 4.7	55.2 ± 2.8
Aslam et al. (2012)	Pakistan	Gypsum	187.7 ± 53.2	8.2 ± 1.9	16.2 ± 3.9
Al-Dadi et al. (2014)	Saudi Arabia	Gypsum	173 ± 2.4	7.7 ± 0.1	3.3 ± 0.1
This study	Albania	CEM I (42.5R)	168.8 ± 24.6	51.2 ± 5.5	16.1 ± 2.3
		CEM II/A-LL (42.5 R)	150.4 ± 19.8	51.0 ± 3.7	16.5 ± 3.6
		CEM II/B-LL (32.5R)	133.7 ± 11.4	46.2 ± 3.6	12.0 ± 3.1
Xhixha et al. (2013b)	Albania	Cement	179.7 ± 48.9	55.0 ± 5.8	17.0 ± 3.3
Tufan and Disci (2013)	Turkey	Cement	147 ± 8	29 ± 5	25 ± 11
Turhan (2008)	Turkey	CEM I	239.0 ± 70.1	29.8 ± 12.3	17.5 ± 6.8
			100.6 ± 28.7	31.7 ± 5.3	15.1 ± 7.4
			172.3 ± 25.4	22.8 ± 9.3	13.8 ± 2.8
		CEM II/A-LL	157.1 ± 50.2	22.4 ± 6.3	12.6 ± 3.3
Khan and Khan (2001)	Pakistan	Portland cement	272.9 ± 67.8	26.1 ± 5.6	28.6 ± 4.3
Papaefthymiou and Gouseti (2008)	Greece	CEM I	154 ± 13	17 ± 1	15 ± 2

Table 2 continued

Author	Country	Material type	⁴⁰ K (Bq kg ⁻¹)	²²⁶ Ra (Bq kg ⁻¹)	²³² Th (Bq kg ⁻¹)
El-Taher et al. (2010)	Egypt	Portland	82.1 ± 4.1	35.6 ± 4.4	43.2 ± 2.2
Aslam et al. (2012)	Pakistan	Portland	295.1 ± 66.9	34.2 ± 11.9	29.1 ± 3.6
Al-Dadi et al. (2014)	Saudi Arabia	Cement	53.1 ± 1.6	65.7 ± 0.5	8.7 ± 0.2

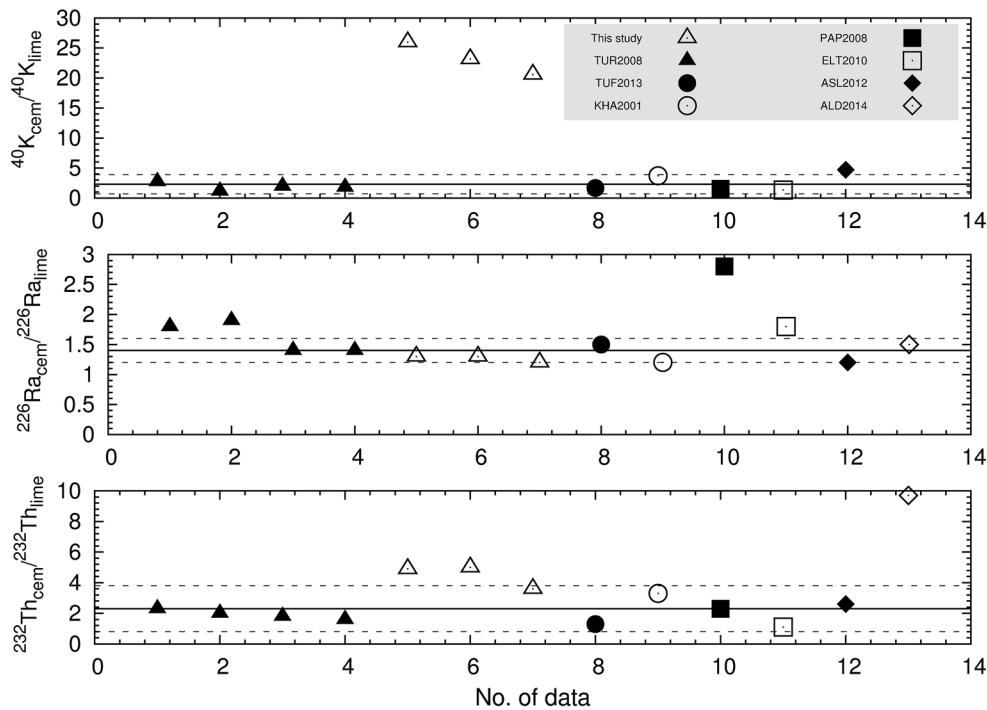


Fig. 1 Activity concentration ratios for ⁴⁰K, ²²⁶Ra and ²³²Th for cement and limestone in different studies. The solid and dashed lines correspond, respectively, to the median and robust standard deviation

where A_{226Ra} , A_{232Th} and A_{40K} are the measured activity concentrations in Bq kg⁻¹ for radium, thorium and potassium, respectively. The activity concentration index applies to building materials and in the case of constituent materials like cements; an appropriate mixing factor needs to be applied. The ACI criterion is recently implemented in Albanian legislation through the Decision of Council of Ministers (V.K.M. Nr. 957 2015) and applies to construction products.

In Fig. 2, we observe that the ACI index in raw materials varies from 0.08 (pyrite) to 0.88 (bauxite), both materials used in relatively low amount. Indeed, the ACI index (0.32) of the clinker depends mainly on limestone and flysch use. Moreover, by increasing the quantity of limestone grinded with gypsum and clinker, from CEM I (42.5R) to CEM II/B-LL (32.5R), we observe a decrease in ACI values. The ACIs for different cement types are found to vary from 0.26 to 0.31 that are below the screening level

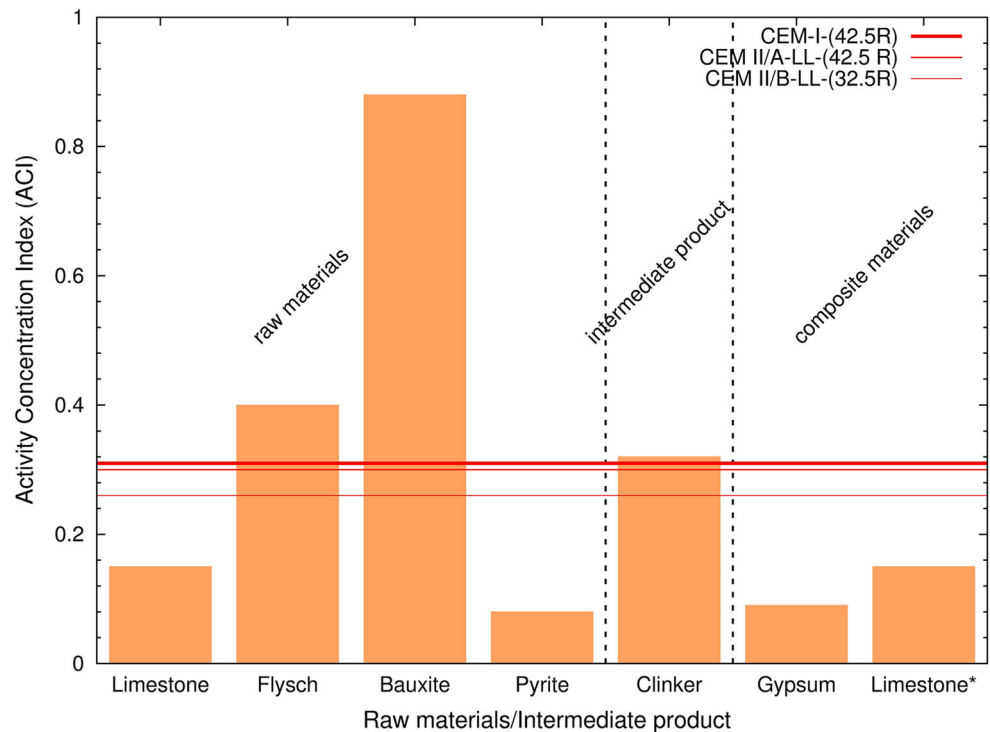
of 1, which roughly indicates that potentially the annual effective dose criterion is less than 1 mSv y⁻¹. These results support the importance of the recommendation of the Council Directive 2013/59/EURATOM (2014) that more attention must be done when screening composite materials, like cement itself. For composite material, the final decision must be applied to building materials, like concrete, mortar or plaster.

The potential contribution to the total external absorbed dose rate (DR in nGy h⁻¹) in the indoor air, due to the presence of natural radionuclides in cements (²²⁶Ra, ²³²Th and ⁴⁰K), is calculated according to UNSCEAR (2000):

$$DR = 0.92 A_{226Ra} + 1.10 A_{232Th} + 0.08 A_{40K} \tag{2}$$

where A_{226Ra} , A_{232Th} and A_{40K} are the measured activity concentrations in Bq kg⁻¹ for radium, thorium and potassium, respectively. The received DR due to the use of cements as building materials is found to vary from 66 to 78 nGy h⁻¹. These values are comparable with the

Fig. 2 Activity concentration index calculated for raw materials, intermediate products, composite materials and different cement types manufactured in Albania



published world average dose rate of 84 nGy h^{-1} (UNSCEAR 2000).

The DR values are used to estimate the annual effective dose rate (EDR in mSv y^{-1}), considering that the population spent, on average, 80% of their time indoors, and using a conversion coefficient for the absorbed doses in the air to the effective dose received by an adult of 0.7 Sv Gy^{-1} (UNSCEAR 2000). The EDR is calculated according to the formula:

$$\text{EDR} = (\text{DR} \times 10^6) \times 0.7 \times (8760 \times 0.8) \quad (3)$$

The corresponding average EDR received due to the use of cements as building materials varies from 0.33 to 0.38 mSv y^{-1} . These results are comparable with the world average annual effective dose rate of 0.41 mSv y^{-1} (UNSCEAR 2000). The EDR values confirm the conclusions derived by applying the ACI screening tool (i.e., the annual effective dose rate is less than 1 mSv y^{-1}) and give a rough estimate of the potential contribution to the annual effective dose rate due to the use of cements as building materials.

Conclusions

The presence of natural radionuclides (^{226}Ra , ^{232}Th and ^{40}K) in Portland cement and Portland-limestone cements is investigated by studying raw materials and intermediate product (clinker). Nine different materials are measured by

means of 45 samples by using gamma-ray spectrometry technique. These results give important indications on the presence of natural radioactivity in cement, where limestone is reasonably the principal contribution to the activity concentration of ^{226}Ra in cement (approximately 70%) and aluminosilicate raw materials are instead the principal contributors for the presence of ^{232}Th and ^{40}K in cement. These conclusions give important information on the radiological assessment in the cement industry, especially in the cases when new quarries are exploited. The activity concentration in different cement types is found to be higher in Portland cements and lowest in Portland-limestone cements, respectively, for ^{226}Ra ($46.2 \pm 3.6\text{--}51.2 \pm 5.5 \text{ Bq kg}^{-1}$), ^{232}Th ($12.0 \pm 3.1\text{--}16.1 \pm 2.3 \text{ Bq kg}^{-1}$) and ^{40}K ($133.7 \pm 11.4\text{--}168.8 \pm 24.6 \text{ Bq kg}^{-1}$). These results are found to be comparable with available bibliographic studies.

From the radiological point of view, the results are used to assess the radiological hazard by means of the activity concentration index. The ACI values are found to vary from 0.26 to 0.31 , which is below the screening level of 1 . A rough estimation of the annual effective dose rate received due to the use of cements as building materials indicates contribution varying from 0.33 to 0.38 mSv y^{-1} . Therefore, based on the ACI values according to the national legislation, these types of cements do not pose any significant risk to humans due to their use in dwellings. However, a more accurate evaluation must be performed on building materials.

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