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Activity Concentration Index Values for Concrete Multistory Residences in Greece Due to Fly Ash Addition in Cement

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Abstract: According to 2013/59/Euratom Directive, the activity concentration index (*ACI*) is required to be estimated for each building material that is of concern from a radiation protection point of view. This index applies to building materials and not to constituents that cannot be used as building materials themselves. Fly ash is a byproduct of coal-fired power plants and is one of the main constituents of cement. The radioactivity in fly ash that is produced by Greek lignite power plants cannot be considered insignificant. For example, in the case of the Megalopolis power plant, the concentration for radioisotopes of the ^{226}Ra chain is found to be about 1 kBq/kg. Since natural radionuclide concentrations, which are harmful to human health in terms of radiation exposure, exist in fly ash, *ACI* should be assessed for building materials containing fly ash. The present study evaluates the *ACI* of concrete containing fly ash cement when used in multistory residential buildings. Results showed that cement produced in Greece by the three main Greek cement production plants, containing lignite fly ash, and used as a material for concrete multistory constructions, should not be considered as "of concern from a radiation protection point of view". Each country that wishes to evaluate the use of fly ash into constructions should repeat the method for the *ACI* uncertainty budget proposed in this study, to assess whether it significantly exceeds the reference value (whether it is of concern from a radiation protection point of view).

Keywords: fly ash addition in cement; concrete multistory residence; activity concentration index; *ACI*

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1. Introduction

Fly ash is a byproduct of coal-fired power plants. Such plants that are established in Greece operate with lignite as their fuel. Lignite in Greece comes from (a) Ptolemais-wide area, but also from even wider territories within the Prefecture of Kozani, Region of Western Macedonia, (b) territories close to Megalopolis town in Peloponnese and (c) territories close to Florina town, also located in the Region of Western Macedonia. Fly ash is used in the cement production process, to a proportion that has been specified by Greek legislation since 1980 [1–3].

Fly ash is an aluminosilicate material, and its chemical/mineral composition depends on the composition of the coal and the combustion conditions. Because of its fineness and pozzolanic nature, fly ash is widely accepted and specified as mineral admixture both in cement and concrete. In concrete, fly ash substitutes a part of cement. Fly ash was recognized as a pozzolanic constituent for use in concrete in 1914. However, research on the material began in 1937. Since then, extensive research has been conducted throughout the world [4]. Fly ash increases concrete workability, reduces water demand for the same slump, increases strength at later curing ages and improves corrosion resistance. It is highly heterogenous, from its particle size to the chemical composition. It can be seen from the several studies in the past that the properties of the final product are dependent on the properties of fly ash. Therefore, it is difficult to arrive at a definitive conclusion on the effect of fly ash on concrete. However, the results available from the extensive studies can be considered as guidelines for further research [5].

Blended fly ash cement can be produced either by intergrinding fly ash with Portland cement clinker or by blending dry fly ash with Portland cement. European Standard EN 197 determines the requirements for fly ash and the composition (percentage by mass) of cement types containing fly ash. The benefits of fly ash blends are that they are cheaper and reduce the amount of clinker needed. This reduction in needed clinker results in the reduction of needed energy and the reduction of the amount of carbon dioxide released into the atmosphere during the production of clinker. Environmental benefits also include the utilization of a waste material and the use of less raw resources. On the other hand, blended fly ash cement has slower setting time, which means its early strength is lower than cement without fly ash, making it unsuitable for use in the precast industry and potentially increasing construction times. It also requires more curing, while its resistance to carbonation is lower, risking corrosion of steel reinforcement. Moreover, with coal-fired power becoming increasingly unpopular (at least in Europe), because of its environmental impact and competition from alternative power sources, such as renewables, the long-term availability of fly ash is in question.

Naturally occurring radioactivity concentrations in lignite used in Thermal Power Plants (TPP) in Greece and the produced fly ash and bottom ash have already been determined since the 1980s [6–8]. These concentrations are generally high, especially in the case of lignite coming from mines in the Megalopolis Peloponnese area, where the concentration for radioisotopes of the ^{226}Ra chain is found to be about 1 kBq/kg [6–12]. As has been already discussed [8], there have been significant differences in the results of various studies relative to the concentration of naturally occurring radioisotopes in lignite-produced fly ash. This could be attributed to differences between the sampling approaches followed in each study. In the same context, the following parameters also contribute:

- Intrinsic variation by time of the under-study concentrations, according to which was the exact deposit where the finally burned lignite was originating from.
- Variations among technical characteristics of the different burning processes.

Article 75 of the European Directive 2013/59/Euratom ([13], called the Directive for the rest of this study) defines the activity concentration index (*ACI*), abbreviated as I for index. This Directive has been adopted by the Greek legislation through article 75 of the Greek Radiation Protection Regulation (Presidential Decree 101/2018, Governments Gazette 194A/20 November 2018, called the Greek Regulation for the rest of this study). Since then, the estimation of *ACI* is obligatory for each "construction material" which is "identified by the Member State (Greece in this case) as being of concern from a radiation protection point of view". The *ACI* formula shown in Equation (1) is provided in Annex VIII of the Directive and adopted in Annex VIII of the Greek Regulation.

$$I = ACI = \frac{C_{Ra-226}}{300\ \text{Bq/kg}} + \frac{C_{Th-232}}{200\ \text{Bq/kg}} + \frac{C_{K-40}}{2000\ \text{Bq/kg}} \tag{1}$$

where C_x is the specific (by mass) activity for x contributor, namely the ^{226}Ra series, the ^{232}Th series and ^{40}K, expressed in Bq/kg.

Essentially, the exposure to ionizing radiation for a person standing inside a structure is an effect of many contributing factors. A similar index has been proposed in Annex A of the 1993 UNSCEAR Report [14]. It aimed at assessing the level of exposure to the gamma radiation field for a person standing on the ground, due to the presence of Naturally Occurring Radioactive Materials (NORM, Figure 1). The UNSCEAR 1993 proposal included the results of the NORMs measurement in Greece [14,15]. As can be seen in Figure 1, the geometry of the person's exposure, when assuming exposure in a standing position on the ground, is relatively simple. The geometry in question is simpler if the soil material is homogeneous. In the same UNSCEAR report, the same index was introduced for assessing personal exposure in indoor spaces, too. The main assumption for this generalization is that the source of the gamma radiation field is construction materials.

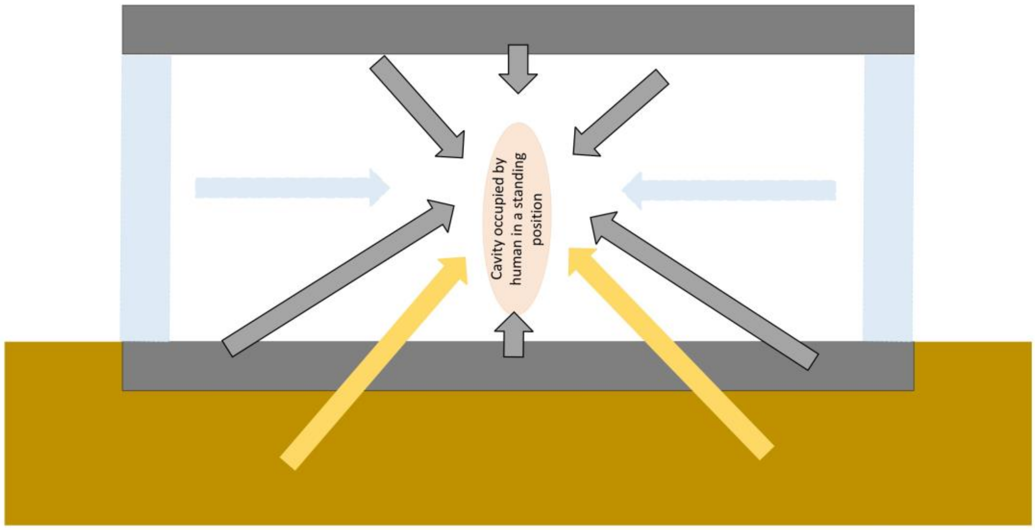

Figure 1. Exposure of a person to gamma radiation field when standing on the ground, due to gamma radiation emitted from soil (yellow arrows) or standing on the floor of a multistory building with concrete as the main structural element, due to gamma radiation emitted by the radioactivity contained in the surrounding materials (dark grey arrows for radiation produced by the floor and the ceiling concrete slabs and light grey arrows for radiation produced by any perpendicular construction materials' masses, e.g. masonry).

Many previous studies estimated *ACI* (or I_γ index, as mentioned in some of them) or other similar indexes such as the ^{226}Ra equivalent activity concentration (Ra_{eq} in Bq/kg) and the external hazard index (H_{ex}, dimensionless as per *ACI*), when gamma radiation is assessed for concrete or even its constituents [16–27]. None of them have proposed a method according to the ISO approach to estimate the uncertainty of the results. Therefore, a producer of construction products/materials or a laboratory contributing to quality control, are not effectively supported to establish a decision rule on whether the product/material is "of concern from a radiation protection point of view".

A certain previous study provides a typology for estimating uncertainty for NORMs measurement in construction materials, according to the ISO methodology [28]. The latter provides a methodology for estimating reliability intervals following measurements on specific specimens that could be either concrete, fresh or hardened, or even samples from concrete constituents. This is a very good approach when establishing a performance indicator, useful for Factory Production Control (FPC). A weakness is that in this way we cannot reveal an overall figure for the entire production or even worse for all production and construction in a country-wide area.

The present study focuses on the case of residences that are part of a multistory concrete construction. Such residential buildings are common in Greece, especially since the 1960s. The exposure geometry of a person standing in the middle of such an indoor space is more complicated than in the case of a person standing on the open field, on plane soil ground. The gamma radiation field is created by concrete, either load bearing or infill, masonry, covering materials such as tiles or plaster, etc., to the extent that they have been used in construction (Figure 1).

In addition, the person is also exposed to the material of the overlying slab, whether it is the floor of the upper floor or the roof of the structure. Also, the person is exposed to the gamma radiation field produced by any vertical structural elements, in a distance that varies significantly by case (Figure 1).

For such complicated exposure geometry, the 1993 UNSCEAR report suggests that *ACI* should be weighed for the mass proportion of the building materials present in the analyzed construction. Equation (2) may be used for estimating the concentration for each of the NORMs that contribute to Equation (1). In this study, where the case is any dwelling in a multistory concrete structure, the activity concentration of NORMs in concrete is the prevailing source external gamma radiation field. Consequently, the result that is assessed in this study is an assessment of the *ACI* only for concrete, so Equation (2) is not used for further incorporation of other building materials (e.g., masonry or tiles).

$$C_x = \frac{\sum_{i=1}^{n} a_i \cdot C_{x,i}}{\sum_{i=1}^{n} a_i} \tag{2}$$

As known [29], the intensity of the external (to the exposed person) gamma field attenuates according to the inverse square distance to the source. This is not incorporated in Equation (2). Essentially, all construction materials are assumed to be homogeneously distributed in the ground on which the person stands. Consequently, to answer the question whether a construction material is "of concern from a radiation protection point of view", we need to consider the distance between the person and the material. On this basis, it is not practical to apply Equation (2) for construction materials that are located too far away from the person, especially if other materials (e.g., a wall) are in the intermediate space. This means that Equation (2) should only be applied for consequent residential spaces (e.g., one single separate room).

On the other hand, an average person uses multiple residential spaces inside an apartment, according to the use of each room. Generally, different rooms inside the same apartment use different construction materials (e.g., a greater mass of tiles is installed on the internal surfaces of a bathroom). A realistic assessment of the person's exposure to the gamma field emitted by the construction materials in the apartment that he inhabits must consider the distribution of time that the person spends in each room.

An assessment of personal exposure to radioactivity contained in the construction products that are produced in Greece has been performed [30,31] considering, in some cases, even the subsequent exposure to radon concentrations [32–34]. It is recalled that in the case of exposure of a person inside an enclosed space, such as inside a residence in a multistory structure, an important source of internal exposure of the person to radiation comes from radon. Therefore, the estimation of the exposure level of the individual inside a residential building is highly complex. It depends on many factors, not only the concentration of radioactivity in the building materials of the structure, but also on the way the resident uses the apartment, such as the mechanical and natural ventilation.

The exact assessment of the *ACI* for the case of a person inside an apartment in a multistory building requires knowledge in relation to all the construction materials that make up the construction, such as the presence of masonry which is also mentioned as a potentially significant source of gamma radiation [35]. This assessment becomes even more complicated if unusual building materials that are likely to contain a high concentration of NORMs have been used in the construction, as, for example, in the case where certain types of granite are used to cover surfaces or as benches. It is also noted that even in cases where the concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the granites may not be particularly high, the presence of other radioisotopes in this material is also mentioned, increasing the level of the produced gamma radiation field [36]. In these cases, the fact that the *ACI* formula (Equation (1)) cannot incorporate the effect from these radioisotopes should also be considered.

If *ACI* estimation aims at an epidemiological risk assessment due to the individual's exposure to gamma radiation coming from the building materials, there are many factors that must be considered. According to Directive 2013/59/Euratom that has been adopted through the Greek Regulation, and specifically in para. 1 of article 75, "The reference level applying to indoor external exposure to gamma radiation emitted by building materials, in addition to outdoor external exposure, shall be 1 mSv per year". According to Annex

VIII of the same Directive, "The activity concentration index value of 1 can be used as a conservative screening tool for identifying materials that may cause the reference level laid down in Article 75(1) to be exceeded". In this case however, definitional uncertainty, which in paragraph 2.27 of ISO VIM [37] is defined as "component of measurement uncertainty resulting from the finite amount of detail in the definition of a measurand", should be considered. In cases of in situ measurements, not the analysis of samples under strict laboratory conditions, definitional uncertainty can be significant [38]. For reasons analyzed above, the question arises as to whether the *ACI* can fulfill the purpose of assessing the individual's exposure, even if the question is restricted to indoor external exposure to gamma radiation emitted by building materials.

Moreover, due to current legislative status in Greece that is compliant with the European Union's New Legislative Framework, any construction product used in Greece shall both comply with the Construction Products Regulation [39] and Greek Radiation Protection Regulation (adopting the European Directive 2013/59/Euratom). Thus, all construction products that are subject to trade in Greece shall be monitored for their performance on *ACI* values. The above framework becomes particularly interesting when building materials are subject to certification procedures.

This paper aims to assess whether the use of lignite fly ash as an additive in cement produced in Greece should be "of concern from a radiation protection point of view". This assessment is applied to concrete produced in Greece using fly ash cement as a constituent, considering that concrete is the predominant construction material (product) for multistory residential buildings. The evaluation of *ACI* values on an approximately 95% confidence level is presented, based on a corresponding uncertainty budget according to ISO GUM methodology [40]. The *ACI* calculation formula (Equation (1)) was applied for concrete mix designs that are possibly used in concurrent constructions in Greece. The estimates are provided assuming that legislative restrictions on the use of fly ash and the requirements of the Greek Concrete Technology Regulation [41] have been met.

Besides with the above assessment for the Greek case of concrete containing cement with lignite fly ash, this paper proposes for the first time a general simple approach, that could be used by any responsible authority that wants to assess whether a construction product is of concern from a radiation protection point of view. Each country that wishes to evaluate the use of fly ash in constructions, could repeat the method for the *ACI* uncertainty budget proposed in this study and apply the same or equivalent decision rule.

2. Data and Methods

Strictly analyzing the uncertainty of the result when applying Equation (1), the contribution to *ACI* estimation parameters is graphically analyzed through a fishbone diagram in Figure 2. This figure shows that an effective uncertainty budget on the usage of Equation (1) does not rely just on the uncertainty budget on the estimation of activity concentration for each participating NORM. Equation (1) relies on the three major NORMs (^{226}Ra and ^{232}Th series of radioisotopes and ^{40}K) concentration but also on a definition uncertainty caused by a lack of knowledge on the exact placement of the construction materials in the residence.

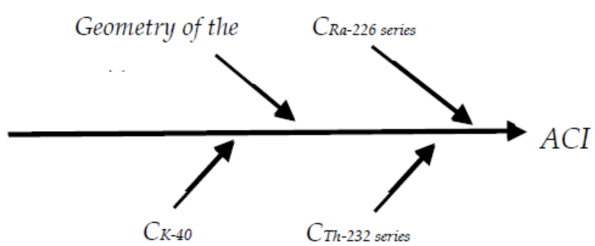

Figure 2. Fishbone diagram of parameters contributing to the estimation of *ACI* when the 2013/59/Euratom Directive formula is used.

As mentioned before, *ACI* estimation should be performed for each individual construction material that is used in the under-assessment structure (Figure 3), but the following analysis focuses only on estimating confidence interval and on applying a decision rule on the estimated *ACI* value only for concrete.

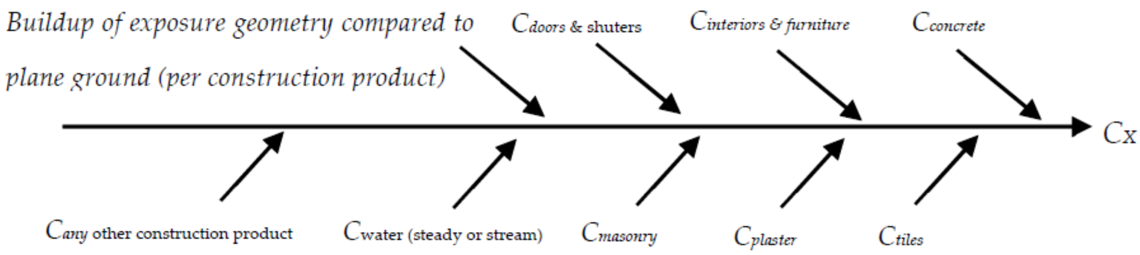

Figure 3. Fishbone diagram showing that the activity concentration that influences the inhabitant of a building is related to many types of construction products (materials).

The presence of fly ash in a concrete construction (i.e., concrete is used as the predominant material) is indirectly affected by the corresponding regulatory context. The status of restriction in the use of fly ash as a cement constituent in Greece could be summarized on a time scale as follows:

Before 1980: There is no existing regulation for the fly ash usage in cement. There is only regulation for natural pozzolana [1].

After 1980: There are two categories of cement containing pozzolana (natural and man-made). In the first category the percentage of pozzolana is determined by the insoluble residue of the cement, which must be 20% maximum. In the second, the insoluble residue of the cement must be 20–40% [2].

After 2002: European Standard EN 197-1 is obligatory, meaning the insoluble residue of the cement must be 35% maximum [3]

Table 1 presents data obtained from the official websites of the three main cement producers in Greece. Those are HERACLES General Cement Company S.A., TITAN Cement Company S.A. and HALYPS Building Materials S.A. In this table, reference is made to the percentage of the presence of cement components, among which is fly ash. The percentage of fly ash present in cement produced in Greece ranges from 0% to 20%. This interval is used as input data for estimating the mass ratio (proportion into cement) in Equation (4) and the corresponding uncertainty budget.

Table 1. Types of cement provided in Greek constructions according to the use of fly ash.

Indicative Types of Cement That the Producer Provides in Year 2023	Cement Producer	Commenting on the Presence of Fly Ash in Cement (As Product)
CEM II/B-M (P-W-L) 32.5N	HERACLES	Natural pozzolana—calcareous fly ash—limestone: 21–35%
CEM II/B-M(W-P-LL) 32.5N	TITAN	Natural pozzolana—calcareous fly ash—limestone: 21–35%
CEM IV/B(P-W) 32.5 (sulfate resistant)	HALYPS	Natural pozzolana—calcareous fly ash—limestone: 36–55%

Table 2 presents results obtained from previous papers on the concentrations of NORMs in samples of fly ash produced by Greek TPPs. This type of fly ash is mainly used by Greek cement producers. The values of mass specific activity of fly ash used in cement that are used in Equation (4) are provided by a review of the values presented in Table 2.

Table 2. Data retrieved from previous works on the potential concentrations of naturally occurring radioisotopes (^{226}Ra, ^{232}Th and ^{40}K) in fly ash produced during lignite combustion in Greek TPPs.

Lignite Origin According to Reference	NORM Concentrations [Bq/kg]		
	^{226}Ra	^{232}Th	^{40}K
Megalopolis (Region of Peloponnese) [7–9,42,43]	293–1058	4–89	308–590
Ptolemais and Kardia (Region of Western Macedonia) [7,8]	204–825	<90	162–299
Aliveri (Evia, Region of Central Greece) [7]	257–357	0.6–2.0	-
Cement producers [31,44]	200–1400	34–84	<650
Greek lignite according to all references	200–1400	<90	<650

Table 3 presents a summary of the results obtained from previous work on the concentrations of radioisotopes of natural origin and in particular the ^{226}Ra and ^{232}Th series and ^{40}K in samples from materials which, according to the current Concrete Technology Regulation, constitute the minimum necessary materials so that the mixture can be called concrete. These values are the input data for Equation (3), specifically for estimating the mass specific activity of concrete constituents other than cement containing fly ash.

Table 3. NORM mass specific concentrations in concrete constituents according to papers related construction materials produced in Greece.

What Was Measured?	NORM Concentrations [Bq/kg]		
	^{226}Ra	^{232}Th	^{40}K
Limestone [31]	5–7	4–10	86–128
Pozzolanic materials [31]	34–40	19–53	298–424
Gypsum [31]	5–9	<LoD *	<LoD
Clinker [31]	12–18	5–23	102–180
Cement not containing fly ash [32]	96	22	200
Total of no fly ash cement constituents	5–96	<53	<424
Cement containing fly ash [32]	215–218	11–26	222–330
Cement (in general) [12]	15–218	10–41	32–457
Sand [31]	7–15	<9.9	<60
Aggregates (in general) [12]	3–46	3–56	19–1048
Aggregates [12,31]	3–46	<56	<1048
Water [42]	<2	<2	<100

* LoD: limit of detection.

It should be noted that all concrete constituents contain NORMs, mainly the aggregates, but even water does so in a significantly lower concentration. Common types of concrete dealt with in this paper mainly contain limestone aggregates, in which the concentration of NORMs is generally low (Table 3). It should be noted that construction materials coming from neighboring countries (e.g., cement imported from Turkey) are used regularly in Greek construction projects. The issue of the NORM presence in them has been studied, providing measurement results like or even higher than those performed for Greek construction products. The intervals reported in Table 3 correspond to the minimum and maximum referenced value. This is assumed to provide intervals at a confidence level of approximately 99.7%. In the same context wherever a reference paper reported a confidence interval based on either the standard deviation of the results or the combined standard uncertainty, Table 3 presents intervals based on these statistics, multiplied by three.

In this paper *ACI* values' reliability limits at an approximately 95% level of confidence are calculated for common types of concrete that may be produced in Greece in year 2023. This proposed calculation could also be repeated for any construction period in the past, considering the corresponding concrete compositions. This paper is restricted to common types of concrete. Consequently, cases of (a) concrete prepared with light weight or heavyweight aggregates and (b) high performance concrete (HPC) were not considered.

Considering that common types of concrete contain cement with fly ash (*fa,cem*), aggregates (*agg*) and water (*w*) in proportions resulting from the corresponding composition study and that any other possible material (e.g., superplasticizer) participates with a non-significant mass ratio, the mass specific activity (concentration) in any case of naturally occurring radioisotopes ^{226}Ra, ^{232}Th and ^{40}K, was calculated according to Equation (3), as also presented graphically in Figure 4.

$$C_{con} = \frac{\alpha_{fa,cem} \cdot C_{fa,cem} + \alpha_{agg} \cdot C_{agg} + \alpha_w \cdot C_w}{\sum \alpha_i} \tag{3}$$

where a_i are the proportions of the three concrete constituents, expressed in kg/m^2 of fresh concrete, i.e., exactly as determined according to the concrete composition study.

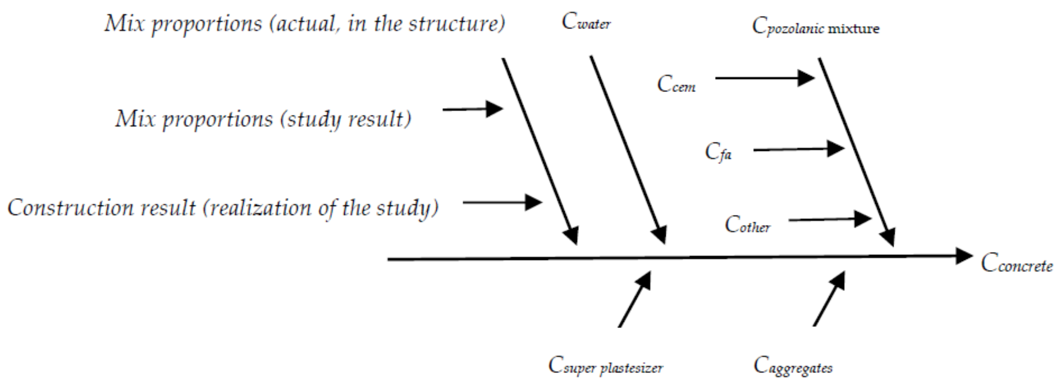

Figure 4. Fishbone diagram showing the parameters that contribute to the concentration of NORMs in a concrete mixture.

In the same equation, no correction has been included according to the possible difference in the moisture content of the fly ash between the values (a) as per the measurement process for determining the activity of NORMs and (b) when mixing with the rest of the cement components. In practice, it was assumed that in these two situations, where the fly ash is expected to be in a dry state, the moisture content is approximately at the same level with no significant difference.

The fresh concrete mix proportions were taken as the ones likely to be used in the Greek construction industry in the year 2022, mainly according to the minimum allowable values resulting from the Greek Concrete Technology Regulation [41] and the maximum possible cement ratio values, considering the economics of residential buildings' construction.

Previous studies were used for estimating the confidence intervals (limits) for mass specific activity values of NORMs in aggregates and water (see Table 2). In the case of the mass specific activity of cement, a calculation was made for each of the three NORMs according to Equation (4):

$$C_{fa,cem} = \alpha_{fa} \cdot C_{fa} + \alpha_{cem} \cdot C_{cem} \tag{4}$$

where a_{fa}, C_{fa} are the mass ratio (proportion into cement) and mass specific activity of fly ash and a_{cem}, C_{cem} are the mass ratio (proportion into cement) and mass specific activity for the rest of the cement constituents.

Previous studies were also used for estimating confidence intervals (limits) for mass specific concentration values of NORMs in cement and fly ash (Table 1). The a_i proportions of cement and fly ash were obtained according to the permissible limits imposed by the concurrent Greek legislation.

As mentioned above, the result of this work, in the form of a confidence interval of about 95% level for the value of *ACI* was obtained through the application of the ISO GUM methodology [40]. Since the information used in the calculations comes from third-party sources, such as legislation limits or results of previous work and measurements, extensive use was made of type B estimates for the combined standard uncertainty of the result.

3. Results

Table 4 provides the uncertainty budget for the estimation of the mass specific activity of NORMs in cement containing lignite fly ash. The uncertainty budget presented in the table is based on the results of Equation (4). As the ratios of fly ash and other cement constituents are linearly correlated (see Equation (4)), the covariance $cov(\alpha_{fa,cem}, \alpha_{cem})$ was assumed based on a linear correlation coefficient equal to −1.

Table 4. Uncertainty budget for the calculation of the confidence interval for the values of the specific activity of ^{226}Ra, ^{232}Th and ^{40}K in cement to which lignite ash has been added.

Factor	Mean Value	Bounds of Possible Values	Distribution of Possible Values	Standard Uncertainty	Sensitivity Coefficient	Contribution to the Uncertainty of the Result
			^{226}Ra			
$\alpha_{fa,Ra226}$	0.1	0.1	Rectangular	0.06	800 Bq/kg	2133 (Bq/kg)^2
$\alpha_{cem,Ra226}$	0.9	0.1	Rectangular	0.06	50 Bq/kg	9 (Bq/kg)^2
$C_{fa,Ra226}$	800 Bq/kg	600 Bq/kg	Triangular	245 Bq/kg	0.1	600 (Bq/kg)^2
$C_{cem,Ra226}$	50 Bq/kg	46 Bq/kg	Triangular	19 Bq/kg	0.9	292 (Bq/kg)^2
		$Cov(\alpha_{fa,Ra226}, \alpha_{cem,Ra226})$				−288 (Bq/kg)^2
$C_{fa,cem,Ra226}$	125 Bq/kg					2746 (Bq/kg)^2
			^{232}Th			
$\alpha_{fa,Th232}$	0.1	0.1	Rectangular	0.06	45 Bq/kg	7 (Bq/kg)^2
$\alpha_{cem,Th232}$	0.9	0.1	Rectangular	0.06	27 Bq/kg	3 (Bq/kg)^2
$C_{fa,Th232}$	45 Bq/kg	45 Bq/kg	Triangular	18 Bq/kg	0.1	3 (Bq/kg)^2
$C_{cem,Th232}$	27 Bq/kg	27 Bq/kg	Triangular	11 Bq/kg	0.9	98 (Bq/kg)^2
		$Cov(\alpha_{fa,Th232}, \alpha_{cem,Th232})$				−9 (Bq/kg)^2
$C_{fa,cem,Th232}$	29 Bq/kg					102 (Bq/kg)^2
			^{40}k			
$\alpha_{fa,K40}$	0.1	0.1	Rectangular	0.06	325 Bq/kg	380 (Bq/kg)^2
$\alpha_{cem,K40}$	0.9	0.1	Rectangular	0.06	212 Bq/kg	162 (Bq/kg)^2
$C_{fa,K40}$	325 Bq/kg	325 Bq/kg	Triangular	133 Bq/kg	0.1	177 (Bq/kg)^2
$C_{cem,K40}$	212 Bq/kg	212 Bq/kg	Triangular	87 Bq/kg	0.9	6130 (Bq/kg)^2
		$Cov(\alpha_{fa,K40}, \alpha_{cem,K40})$				−496 (Bq/kg)^2
$C_{fa,cem,K40}$	223 Bq/kg					6353 (Bq/kg)^2

Table 4 shows the following:

- The proportion of cement constituents is reported as a dimensionless number, meaning the mass fraction of constituents and the value of the bounds of possible values [40] refer to the half interval on both sides of the mean to produce a confidence interval of about 99.7% level.
- It should be noted that the range of values considered for the proportion of fly ash addition to cement is a worst-case scenario compared to what is known for the actual construction activity in Greece in the last 20 years. The proportion of fly ash added into cement has decreased in recent years due to a decrease in the production of fly ash by Greek TPPs. In the early years of this twenty-year period, the proportion of

fly ash added to cement was approximately up to 15% by mass. In current years, the maximum value of this rate has been reduced to approximately 3%. A range of values for $\alpha_{fa,Ra226}$ = 0–0.2 (i.e., 0% to 20% fly ash percentage, by mass, in the produced cement) was assumed. Such an interval is so wide to includes any effect of a possible difference in the moisture content of the fly ash between samples' moisture content when analyzed in a gamma spectroscopy setup for determining ^{226}Ra, ^{232}Th and ^{40}K activity and the moisture content of all other cement constituents, during their mix.

- The uncertainty budget in Table 4 leads to confidence intervals at an approximately 95% level presented in Table 5 due to applying the "law of error propagation" [40]. In this context, the bounds (a range) of possible values and a corresponding distribution is assumed to produce the standard uncertainty for each variable that contributes to Equation (4). Standard uncertainties are multiplied by the corresponding sensitivity coefficients. These are the partial derivatives of the applied equation as per the variables for which the sensitivity coefficient is estimated. The right-most column of Table 4 shows the contribution of each variable to the finally estimated uncertainty of the result for each NORM as the square value of the result of this multiplication or the value of the covariance between the two mass proportions. The results presented in Table 4 are the sum of these contributions, which is the square of the combined standard uncertainty relative to the result of Equation (4), for each NORM.
- Table 5 presents the result of Equation (4) as an approximately 95% confidence interval. These intervals were estimated as the square root of the results obtained from Table 4, for each NORM, multiplied by a coverage factor k equal to 2.
- The values provided in Table 5 are used as input data in calculations provided in Table 6 in relation to the possible values of the mass specific activity of cement containing fly ash. Table 6 provides the uncertainty budget for the estimation of mass specific activity of fresh concrete constituents: cement, aggregate and water. The uncertainty budget presented in these tables refers to Equation (3). It is noted that the ratios of these three concrete constituents have an expected correlation between them, as any decision to increase the ratio of the one, (e.g., of the cement to seek a greater strength in the hardened concrete), leads to a corresponding readjustment of the ratio of at least one or even both the remaining constituents.

Table 5. Estimated 95% confidence intervals for NORM mass specific concentrations (Bq/kg) in cement produced in Greece during the year 2023.

Estimated for	$C_{fa,cem}$ (Bq/kg)
^{226}Ra	$(1.2 \pm 1.0) \times 10^2$
^{232}Th	29 ± 20
^{40}K	$(2.2 \pm 1.6) \times 10^2$

In Table 6, a weak negative correlation (linear correlation coefficient $r = -0.2$) between the cement ratio and aggregate ratio and between the water and aggregate ratio was assumed. An increased linear correlation coefficient, equal to 0.5, was assumed between the ratio of cement and the ratio of water, as it is known that concrete producers put efforts to maintain a steady value for the cement to water ratio in the mixture.

The uncertainty budget in Table 6 leads to confidence intervals at an approximately 95% level presented in Table 7 because of the application of the "law of error propagation" [40], through the same process that was followed for Tables 4 and 5.

According to the above calculations, and by applying Equation (1), it follows that the value of the *ACI* for the common types of concrete used in multistory constructions in the Greek area is 0.47 ± 0.20. The decision rule by which the result is compared to the reference value of EURATOM Directive is that the entire 95% confidence interval for the values of *ACI* should be below 1. Consequently, the maximum value on a 95% confidence interval is

0.67, which means that the entire confidence interval is significantly less than 1 (less than the maximum reference value of EURATOM Directive).

Table 6. Uncertainty budget for the calculation of the confidence interval for the values of the specific activity of ^{226}Ra, ^{232}Th and ^{40}K in fresh concrete.

Factor	Mean Value	Bounds of Possible Values	Distribution of Possible Values	Standard Uncertainty	Sensitivity Coefficient	Contribution to the Uncertainty of the Result
			^{226}Ra			
$\alpha_{fa,cem}$	320 kg/m^2	40	Triangular	16	0.03	0.33 (Bq/kg)^2
α_{agg}	1500 kg/m^2	200	Triangular	82	−0.005	0.16 (Bq/kg)^2
α_w	164 kg/m^2	52	Triangular	21	−0.01	0.1 (Bq/kg)^2
$C_{fa,cem,Ra226}$	125 Bq/kg			42	0.14	35 (Bq/kg)^2
$C_{agg,Ra226}$	24 Bq/kg	22	Triangular	9	0.79	50 (Bq/kg)^2
$C_{w, Ra226}$	2 Bq/kg	2	Triangular	1	0.07	0.003 (Bq/kg)^2
		$Cov(\alpha_{fa,cem}, \alpha_{agg})$ (r = −0.2, is assumed)				0.09 (Bq/kg)^2
		$Cov(\alpha_{fa,cem}, \alpha_w)$ (r = 0.5, is assumed)				−0.18 (Bq/kg)^2
		$Cov(\alpha_{agg}, \alpha_w)$ (r = −0.2, is assumed)				−0.05 (Bq/kg)^2
$C_{con,Ra226}$	35 Bq/kg		-			85 (Bq/kg)^2
			^{232}Th			
$\alpha_{fa,cem}$	320 kg/m^2	40	Triangular	16	0.001	0.0004 (Bq/kg)^2
α_{agg}	1500 kg/m^2	200	Triangular	82	0.0007	0.004 (Bq/kg)^2
α_w	164 kg/m^2	52	Triangular	21	−0.01	0.05 (Bq/kg)^2
$C_{fa,cem,Th232}$	29 Bq/kg			10	0.14	2 (Bq/kg)^2
$C_{agg,Th232}$	28 Bq/kg	28	Triangular	11	0.79	81 (Bq/kg)^2
$C_{w,Th232}$	2 Bq/kg	2	Triangular	0,8	0.07	0.003 (Bq/kg)^2
		$Cov(\alpha_{fa,cem}, \alpha_{agg})$ (r = −0.2, is assumed)				−0.0005 (Bq/kg)^2
		$Cov(\alpha_{fa,cem}, \alpha_w)$ (r = 0.5, is assumed)				−0.004 (Bq/kg)^2
		$Cov(\alpha_{agg}, \alpha_w)$ (r = −0.2, is assumed)				0.006 (Bq/kg)^2
$C_{con,Th232}$	26 Bq/kg		-			83 (Bq/kg)^2
			^{40}k			
$\alpha_{fa,cem}$	320 kg/m^2	40	Triangular	16	−0.10	3 (Bq/kg)^2
α_{agg}	1500 kg/m^2	200	Triangular	82	0.03	7 (Bq/kg)^2
α_w	164 kg/m^2	52	Triangular	21	−0.17	14 (Bq/kg)^2
$C_{fa,cem,K40}$	223 Bq/kg			78	0.14	119 (Bq/kg)^2
$C_{agg,K40}$	524 Bq/kg	524	Triangular	214	0.79	28,422 (Bq/kg)^2
$C_{w,K40}$	50 Bq/kg	50	Triangular	20	0.07	2 (Bq/kg)^2
		$Cov(\alpha_{fa,cem}, \alpha_{agg})$ (r = −0.2, is assumed)				2 (Bq/kg)^2
		$Cov(\alpha_{fa,cem}, \alpha_w)$ (r = 0.5, is assumed)				6 (Bq/kg)^2
		$Cov(\alpha_{agg}, \alpha_w)$ (r = −0.2, is assumed)				4 (Bq/kg)^2
$C_{con,K40}$	448 Bq/kg		-			28,580 (Bq/kg)^2

Table 7. Estimated 95% confidence intervals for NORM mass specific concentrations (Bq/kg) in concrete produced in Greece during the year 2023.

Estimated for	C_{con} (Bq/kg)
^{226}Ra	35 ± 18
^{232}Th	26 ± 18
^{40}K	(4.5 ± 3.4) × 10^2

4. Discussion

Despite the considered addition of fly ash to cement, none of ^{226}Ra, ^{232}Th and ^{40}K emerged as a prevailing radiological factor as compared to others. It should be noted that ^{40}K concentration in common fresh concrete is unlikely to affect *ACI* values. Covariances between the proportions of the concrete constituents are also unlikely to affect *ACI* values. On the other hand, the sensitivity coefficient related to changes in NORMs concentrations in the aggregates is relatively high. The possible addition of aggregates that do not belong to the—usually used in Greece—limestone category, must be considered.

It is emphasized again that this paper is restricted to the common types of concrete used in the load-bearing structure of a multistory building (slabs, columns and beams), in which case the presence of cement is determined: (a) by the minimum limits for the cement content of the concrete due to the required minimum strength and (b) the economy of construction, which, according to the logic of the construction market, sets the respective upper possible limits of the concrete's cement content.

The ratios shown in the calculations in Table 6 correspond to fresh concrete. If a reduction is attempted in the hardened concrete, the loss of water mass due to evaporation during the hardening period should be considered. As already discussed in the introduction of this paper, it is impractical to attempt to accurately reproduce the exposure geometry of the individual residual in the structure, as this is affected by many significant uncertainty factors. The estimation and uncertainty budget presented in this paper provide an index which makes sense for concrete producers by assisting them in assessing the performance of their product, especially in the context of the regulatory control of trade in the European market.

It should be noted that the above estimate for *ACI* is not a result that corresponds to a specific type of concrete or, even more so, an individual mix proportions' study. It is an estimate of the confidence interval at a level of approximately 95% for all common types of concrete that are expected to have been produced in the Greek area within the last twenty years. The most important feature of this interval is the upper value of *ACI*, equal to 0.67 (0.47 + 0.20). It indicates that the common types of concrete containing fly ash cement are impossible to approach the limit value equal to 1.

ACI can be considered from the perspective of monitoring the performance of construction materials, as required for other performances under the EU Construction Products Regulation [39]. In this context, *ACI* with a reference value equal to 1, could be a performance indicator. A same case that has been regulated in the EU is the ranking of buildings according to their energy performance indicator (EPI), which is obtained as the quotient of the estimated energy consumption in a building to a corresponding reference value [45]. The similarities with the *ACI* are many, as the denominators in Equation (1) provide a clear, and horizontal, reference value.

The methodology presented in this paper may be useful, too, for laboratories performing testing on the NORMs concentration in building materials. According to the International Standard ISO/IEC 17025:2017 [46], the conformity assessment of a product subjected to laboratory testing should be carried out based on a prescribed decision rule. This rule must consider the level of uncertainty of the result of the test(s). If a laboratory is asked to give an opinion or interpretation in relation to the compliance of a construction product, such as concrete, with the *ACI* < 1 criterion, then the laboratory should have established an uncertainty budget, like the one presented in this paper.

5. Conclusions

Fly ash used as an additive in cement for concrete multistory constructions in Greece should not be considered as "of concern from a radiation protection point of view", according to the provisions of 2013/59/Euratom Directive.

Each country that wishes to evaluate the use of fly ash in constructions should repeat the method for the *ACI* uncertainty budget proposed in this study, to assess whether it significantly exceeds the value equal to 1.

When *ACI* is estimated for each construction product but also for the entire construction, given the mass proportions of all the used products in a construction, it could serve as a performance indicator. This is useful in construction product certification procedures, with a special interest for concrete producers.

The above-mentioned use of *ACI*, along with the corresponding uncertainty estimation, could be a useful tool for decision making for all the interested parties, such as construction product producers, building designers and testing laboratories that perform measurements focused on estimating *ACI*.

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