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Building Materials Radioactivity in Poland[3]

Radioaktywność materiałów budowlanych w Polsce

Радиоактивность строительных материалов в Польше

ABSTRACT

Introduction: The systematic research of the natural radioactivity of raw and building materials has been conducted in Poland since 1980. Basing on the results of these studies, carried out by both the Central Laboratory for Radiological Protection (CLOR) and over 30 other research laboratories in our country, the national database of natural radioactivity measurements has been set up. The database is supervised by the CLOR and contains the results of the measurements for more than 42 000 samples analysed since 1980 up till now. Due to the economic development of the country, since 1990 there has been an increase in the number of the natural radioactivity measurements of raw and building materials.
Objective: The aim of this article is the presentation and evaluation selected of raw and building materials in terms of radiology.
Method: In Poland the possibility of using different raw and ready building materials is classified due to the value of activity coefficients f_1 and f_2. Activity coefficient f_1 specifies the content of natural isotopes in a test material and is the coefficient of the gamma radiation exposure to the whole body. Activity coefficient f_2 specifies the content of radium ^{226}Ra (mother of isotope ^{222}Rn) in the test material and is the coefficient of the exposure of the lungs epithelium to the alpha radiation emitted by the decay products of radon, breathed into with air by the human respiratory system. Activity coefficients are described by the natural radioactivity of potassium ^{40}K, radium ^{226}Ra and thorium ^{228}Th. Activity concentration of these radionuclides is determined by the MAZAR analyser with a scintillation detector. It is a three-window analyser, which measures samples in the range from 1.26 MeV to 2.85 MeV.
Results: This paper shows the values of activity coefficients f_1 and f_2 for a few selected raw and building materials like ash, concrete, cement and ceramics. Additionally, activity coefficients f_1 and f_2 for carbon are discussed. Carbon, as a precursor to a few building raws, (ash, slag, mixture of ash and slag) has been measured in significant amounts since 1996. Average value of its activity coefficient f_1 was between 0.15 and 0.43 while an average index f_2 was from 14.7 Bq/kg to 44.2 Bq/kg for results collected in 1980-2012. Average values of activity coefficients f_1 and f_2 for carbon are the lowest of all measured and compared materials described in this paper. Average value of activity coefficient f_1 of ash as a by-product of coal combustion is a few times higher than for carbon and is higher than the limit value equals 1.0 for results from almost all years.
Conclusions: In the paper, average value and range of dose rate for these several raw and building materials have been shown. An average dose rate is between 31.8 nGy/h for carbon up to 140.8 nGy/h for ash.

Keywords: building materials, natural radioactivity, ^{40}K, ^{226}Ra, ^{228}Th
Type of article: original scientific article

ABSTRAKT

Wprowadzenie: Od 1980 roku w Polsce prowadzone są systematyczne badania naturalnej promieniotwórczości surowców i materiałów budowlanych. W oparciu o wyniki badań m.in. Centralnego Laboratorium Ochrony Radiologicznej (CLOR) oraz ponad 30 innych laboratoriów badawczych w naszym kraju powstała ogólnopolska baza pomiarów promieniotwórczości naturalnej. Baza ta jest nadzorowana przez CLOR i zawiera wyniki pomiarów dla ponad 42 000 zbadanych próbek od 1980 roku do chwili obecnej. W związku z rozwojem gospodarczym kraju, od 1990 roku nastąpił wzrost liczby pomiarów naturalnej radioaktywności surowców i materiałów budowlanych.
Cel: Celem artykułu jest przedstawienie i ocena wybranych surowców i materiałów budowlanych pod względem radiologicznym.
Metoda: W Polsce możliwość wykorzystania różnych surowców i gotowych materiałów budowlanych uzależniona jest od wartości wskaźników aktywności f_1 i f_2. Wskaźnik aktywności f_1 określa zawartość naturalnych izotopów w badanym materiale i jest współczynnikiem narażenia całego ciała na promieniowanie gamma. Wskaźnik aktywności f_2 określa zawartość radu ^{226}Ra w badanym materiale i jest wskaźnikiem narażenia nabłonka płuc na promieniowanie alfa emitowane przez produkty rozpadu radonu, pobrane wraz z powietrzem przez układ oddechowy człowieka. Wskaźniki aktywności opisane są przez promieniotwórczość naturalną potasu ^{40}K, radu ^{226}Ra i toru ^{228}Th. Stężenie aktywności tych

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radionuklidów jest określana za pomocą analizatora MAZAR z detektorem scyntylacyjnym. Analizator jest trzyzakresowy. Mierzy próbki w zakresach 1,26 do 1,65 MeV, od 1,65 do 2,30 MeV i od 2,30 do 2,85 MeV.

Wyniki: W artykule przedstawiono wartości wskaźników aktywności f_1 i f_2 dla kilku wybranych surowców i materiałów budowlanych, takich jak popiół, beton, cement i ceramika. Dodatkowo omówiono wskaźniki aktywności f_1 i f_2 dla węgla. Węgiel jako prekursor kilku surowców budowlanych (popiół, żużel, mieszanina popiołowo-żużlowa) mierzono w znaczących ilościach od 1996 roku. Średnia wartość jego wskaźnika aktywności f_1 wynosiła od 0,15 do 0,43, podczas gdy średni wskaźnik f_2 od 14,7 Bq/kg do 44,2 Bq/kg dla wyników zebranych w latach 1980-2012. Średnie wartości wskaźników aktywności f_1 i f_2 węgla są najniższe spośród wszystkich zmierzonych i porównanych materiałów opisanych w niniejszym artykule. Średnia wartość wskaźnika aktywności f_1 popiołu, jako produktu ubocznego spalania węgla, jest kilka razy wyższa niż w przypadku węgla i jest wyższa od wartości granicznej równej 1,0 w wynikach z prawie wszystkich lat.

Wnioski: W artykule przedstawiono wartość średnią i zakres mocy dawki dla tych kilku wybranych surowców i materiałów budowlanych. Średnia moc dawki wynosi od 31,8 nGy/h w przypadku węgla do 140,8 nGy/h w przypadku popiołu.

Słowa kluczowe: materiały budowlane, promieniotwórczość naturalna, ^{40}K, ^{226}Ra, ^{228}Th
Typ artykułu: oryginalny artykuł naukowy

АННОТАЦИЯ

Введение: С 1980 года в Польше проводятся систематические исследования естественной радиоактивности сырья и строительных материалов. На основании результатов этих исследований, проводимых как Центральной лабораторией радиологической защиты (CLOR), так и более 30 другими научно-исследовательскими лабораториями в нашей стране была создана общенациональная база данных измерений естественной радиоактивности. Эта база данных находится под контролем CLOR и содержит результаты измерений более 42 000 образцов, проанализированных с 1980 по настоящее время.

В связи с экономическим развитием страны с 1990 года наблюдается увеличение числа измерений естественной радиоактивности сырья и строительных материалов.

Цель: Цель данной статьи состоит в представлении и оценке выбранного сырья и строительных материалов с точки зрения их радиологических свойств.

Метод: В Польше использование различных сырьевых материалов и готовых строительных материалов классифицируется относительно значений показателей активности f_1 и f_2. Показатель активности f_1 определяет содержание природных изотопов в исследуемом материале и является фактором опасного воздействия гамма-излучения на целое тело). Показатель активности f_2 определяет содержание радия ^{226}Ra в исследуемом материале и является показателем опасного воздействия на альвеолы легких, вызванного альфа-излучением, эмитированного продуктами распада радия, которые поступают вместе с воздухом в дыхательную систему человека. Показатели активности описываются естественной радиоактивностью калия ^{40}K, радия ^{226}Ra и тория ^{228}Th. Концентрация активности этих радионуклидов определяется при помощи анализатора MAZAR со сцинтилляционным детектором. Анализатор работает в трех диапазонах и измеряет образцы для значений от 1,26 до 1,65 MeV, от 1,65 до 2,30 MeV и от 2,30 до 2,85 MeV.

Результаты: В статье представлены показатели активности f_1 и f_2 для выбранного сырья и строительных материалов, таких как зола, бетон, цемент и керамика. Кроме того, в статье обсуждались показатели активности f_1 и f_2 угля. Уголь, который являлся предшественником некоторых строительных материалов (золы, шлака, смеси золы и шлака), измеряли достаточно часто начиная с 1996 года. Среднее значение показателя активности f_1 колебалось от 0,15 до 0,43, в то время, как средний показатель f_2 составял 14,7 Бк/кг до 44,2 Бк/кг для результатов, полученных в 1980-2012 годах. Средние значения показателей активности f_1 и f_2 угля являются самыми низкими из всех измеренных и сравниваемых материалов, описанных в этой статье. Среднее значение активности f_1 золы в качестве побочного продукта от сжигания угля в несколько раз выше, чем угля, и выше, чем предельное значение 1,0 для результатов, собранных за все годы исследований.

Выводы: В статье представлено среднее значение и диапазон мощности дозы для нескольких типов сырья и строительных материалов. Средняя мощность дозы колеблется от 31,8 нГр/ч в случае угля, и до 140,8 нГр/ч для золы.

Ключевые слова: строительные материалы, естественная радиоактивность, ^{40}K, ^{226}Ra, ^{228}Th
Вид статьи: оригинальная научная статья

1. Introduction

At present, the evaluation of raw and building materials in terms of the presence of radioactivity concentration shall apply to the criteria published in the Ordinance of the Council of Ministers of 2 January 2007, (Journal of Laws No 4 pos. 29) "on the requirements regarding the content of natural isotopes of ^{40}K, ^{226}Ra and ^{228}Th in the raw materials and the materials used in buildings intended for the dwelling of people and livestock, and also in the industrial waste used in buildings and control of the content of these isotopes". It is the primary and the only act specifying the requirements for raw and building materials used in various types of construction activities.

The regulation classifies the possibility of applying different raw and building materials in various types of housing by specifying two parameters [1]:

- the qualification coefficient f_1 – specifies the content of natural isotopes in the test material and is the coefficient of the gamma radiation exposure to the whole body,
- the qualification coefficient f_2 (concentration of ^{226}Ra) – specifies the content of the test material and is the coefficient of the exposure of the lungs epithelium to the radiation emitted by the decay products of radon, i.e. alpha particles breathed into with air by the human respiratory system.

The values of the activity coefficients f_1 and f_2 are described by the formulae (1) and (2):

$$f_1 = S_K/3000\ [Bq/kg] + S_{Ra}/300\ [Bq/kg] + S_{Th}/200\ [Bq/kg] \quad (1)$$

$$f_2 = S_{Ra}\ [Bq/kg] \quad (2)$$

where: S_K, S_{Ra} and S_{Th} are the values of the activity concentration of the relevant radionuclides in Bq/kg.

Depending on the qualification coefficients, the raw and building materials are licensed to be utilized in various constructions.

The limit values of the activity coefficients f_1 and f_2 for the construction of dwellings are:

$$f_1 = 1 \text{ and } f_2 = 200\ Bq/kg$$

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The measured values of **f_1 and f_2** are being considered during the evaluation, but none can exceed the limit value by more than 20%, i.e.: ***$f_1 \leq 1.2$* and *$f_2 \leq 240$ Bq/kg*.**
The total uncertainty value of the measurements has also been limited to 20% of the value of the coefficients f_1 and f_2, not less than 0.8 of their limit values.

Very important, from the point of view of radiation protection, is an increase in exposure due to natural radiation sources that causes an increase in the annual effective absorbed dose of about 1mSv. The limit values of activity coefficients f_1 and f_2 meet the criterion of not exceeding the annual effective dose which equals 1mSv. Similar limits for housing apply in Norway, while in Lithuania and Finland limits for raw and construction materials relate only to activity index f_1.

The limit values for the remaining applications in the construction industry are:

1. ***$f_1 = 2$ and $f_2 = 400$*** **[Bq/kg]** regarding the industrial waste used in surface objects constructed in the built-up areas, or designed for built-up areas in the local urbanization plans, or for leveling of such areas;
2. ***$f_1 = 3,5$ and $f_2 = 1000$*** **[Bq/kg]** regarding the industrial waste used in surface parts of the objects not mentioned in point 1 and for leveling of the areas not mentioned in the above point;
3. ***$f_1 = 7$ and $f_2 = 2000$*** **[Bq/kg]** regarding the industrial waste used in the underground parts of the objects mentioned in point 2, and the underground constructions, including railway and road tunnels, excluding the industrial waste used in underground mining pits.

The gamma dose rate at the height of 1 m over the unlimited flat surface of the terrain of the average density of r = 1,6g/cm³ is determined semi-empirically by the formula (3) [2]:

$$D = 0{,}043\, S_K + 0{,}43 S_{Ra} + 0{,}66\, S_{Th}\ [nGy/h] \qquad (3)$$

where S_K, S_{Ra}, S_{Th} – activity concentration in [Bq/kg] of respectively ^{40}K, ^{226}Ra (in radioactive equilibrium with daughter nuclides) and ^{228}Th (in radioactive equilibrium with daughter nuclides).

In addition, the absorbed dose rate at 1 m above ground level, road or facility should not exceed 0.3 μGy/h for the application of industrial waste to the levelling and the construction of roads, sports and recreational facilities.

2. Equipment and method

Most laboratories use the MAZAR analysers of the new generation or the AZAR ones of the older generation with the NaI (Tl) detector to determine the qualification coefficients f_1 and f_2 in Poland. These are three-windows analysers which determine the concentration of radioactive ^{40}K, ^{226}Ra and ^{228}Th. The windows of analyser are ^{40}K, ^{226}Ra and ^{228}Th in the ranges of 1.26 MeV to 1.65 MeV, 1.65 MeV to 2.30 MeV and 2.30 MeV to 2.85 MeV respectively. The efficiency calibration of the detector was performed with the use of volume standards ^{40}K, ^{226}Ra and ^{228}Th and a standards matrix (for background). The standard source are Marinelli beakers of 1.5 dm³ volume and they are made for most laboratories by the Central Laboratory for Radiological Protection in Poland. The density of standard sources is equal 1.6 g/cm³, while the density of samples of raw and building materials are within the range 0.6 to 2.0 g/cm³. In order to minimize the outside gamma background, the detector is placed in a shielding house made of 50 mm lead. Shredded and screened through a 2 -millimetres-mesh sieve samples are packed into Marinelli beakers of 1.5 dm³ volume and sealed. The samples are measured after the time elapse to establish the radioactive equilibrium between ^{226}Ra - ^{214}Bi and ^{228}Th - ^{208}Tl after about 2 weeks. All measurements were made in the same geometry and once.

3. The results and discussion

The qualification coefficient f_1 is representative for specifying the content of natural isotopes in the test material, because it contains natural radioactive isotopes, which are mostly in the earth crust such as ^{40}K, ^{226}Ra (represent ^{238}U series) and ^{228}Th (represent ^{232}Th series). Other natural radionuclides have minor significance.

Since 1990, there has been an increase in the number of qualification measurements of the natural radioactivity of raw and building materials, because of the economic development of the country. However, during the recent years the number of the natural radioactivity tests has been reduced, due to the wider usage of the construction materials of foreign origin, which are not subject to the mandatory testing in Poland.

The values of f_1 and f_2 for different raw and building materials (carbon, ash, cement, concrete and ceramics) are shown on Fig. 1 to Fig. 10 as a function of time.

Average values of the activity coefficients f_1 and f_2 for carbon are in the range of 0.15 to 0.43 (Fig. 1) and 14.7 Bq/kg to 44.2 Bq/kg (Fig. 2) respectively. In the whole period of time the values of f_1 and f_2 have not been exceeded. The difference between the minimum and maximum of f_1 and f_2 increases with the number of samples, which may be caused by measurements of materials of various geological origin (place of coal mining).

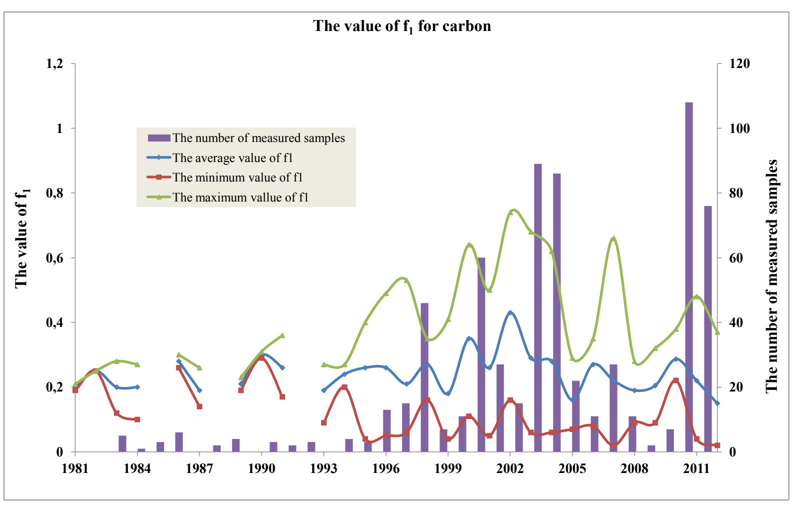

Fig. 1. The value of f_1 for carbon at different times
Source: Own elaboration.

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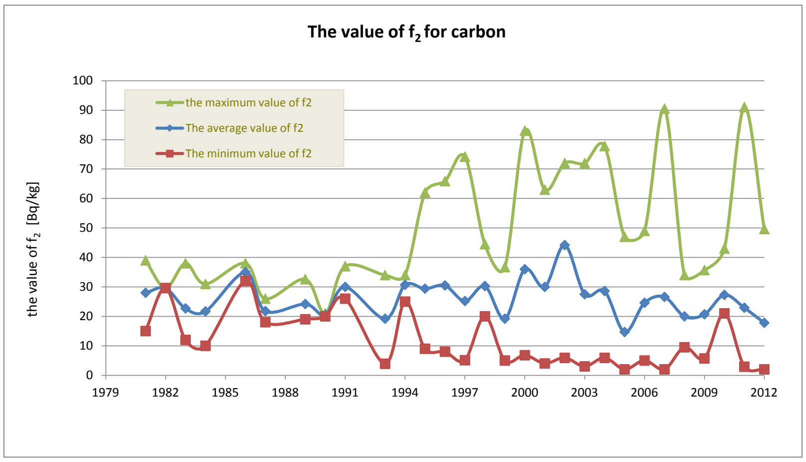

Fig. 2. The value of f_2 for carbon at different times
Source: Own elaboration.

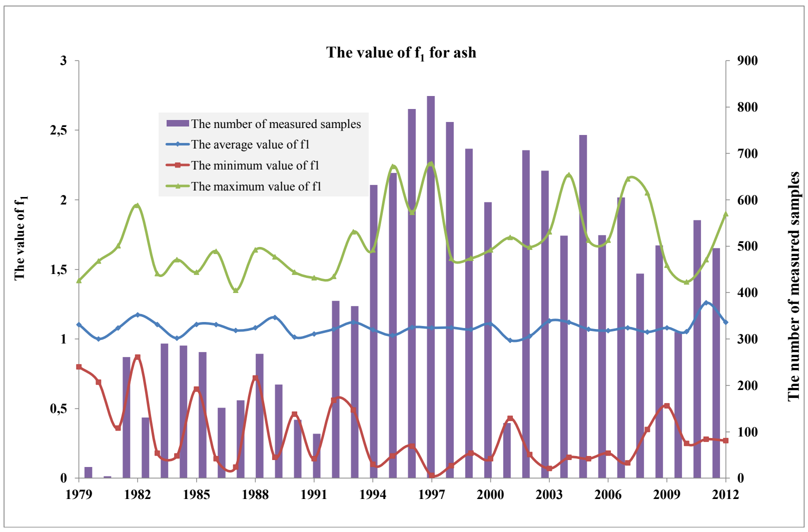

Fig. 3. The value of f_1 for ash at different times
Source: Own elaboration.

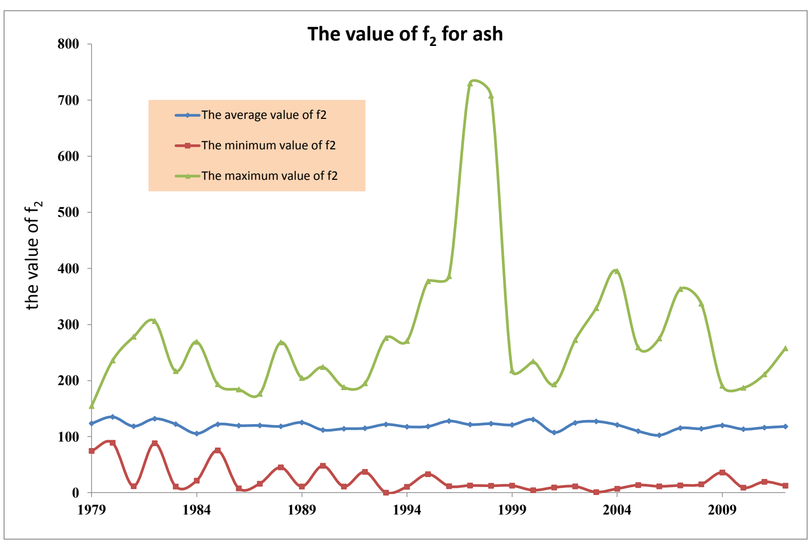

Fig. 4. The value of f_2 for ash at different times
Source: Own elaboration.

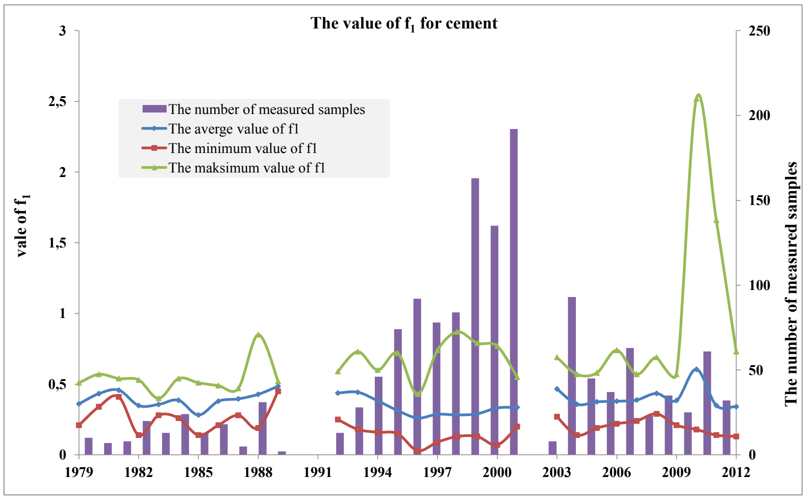

Fig. 5. The value of f_1 for cement at different times
Source: Own elaboration.

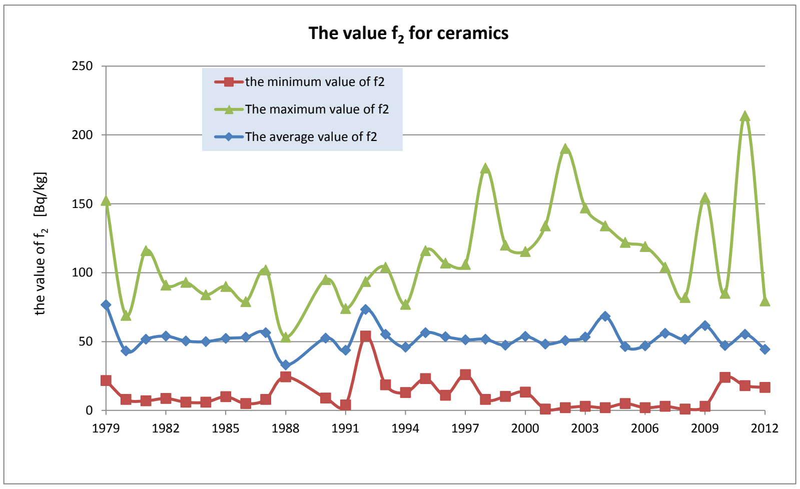

Fig. 6. The value of f_2 for cement at different times
Source: Own elaboration.

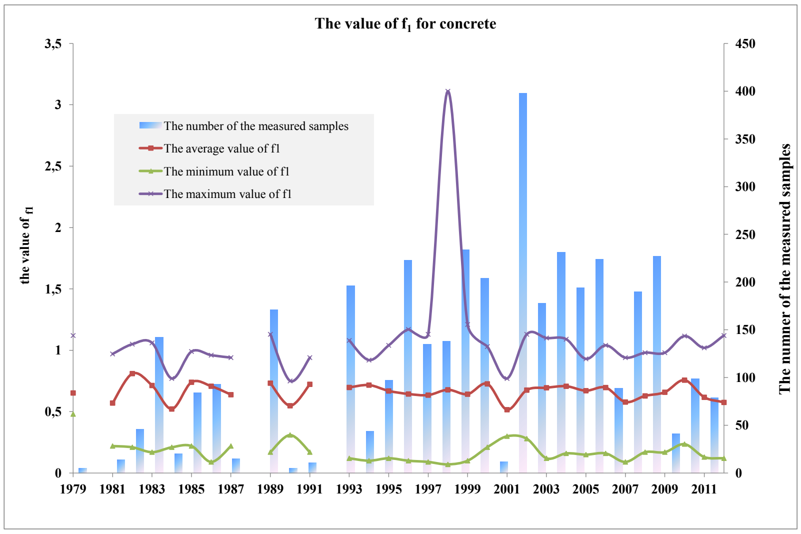

Fig. 7. The value of f_1 for concrete at different times
Source: Own elaboration.

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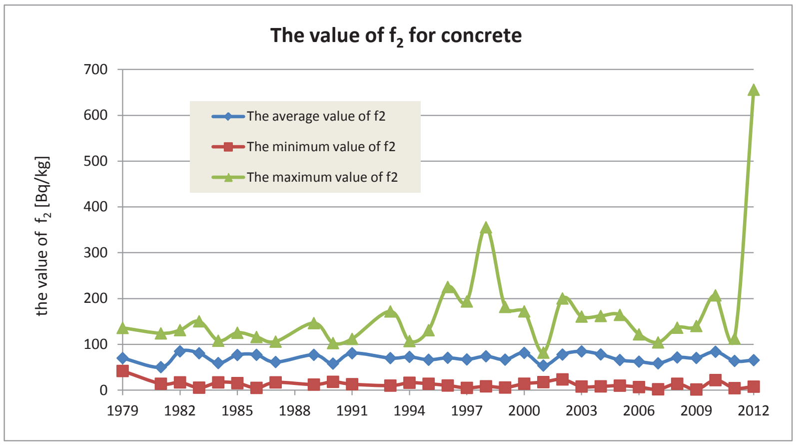

Fig. 8. The value of f_2 for concrete at different times
Source: Own elaboration.

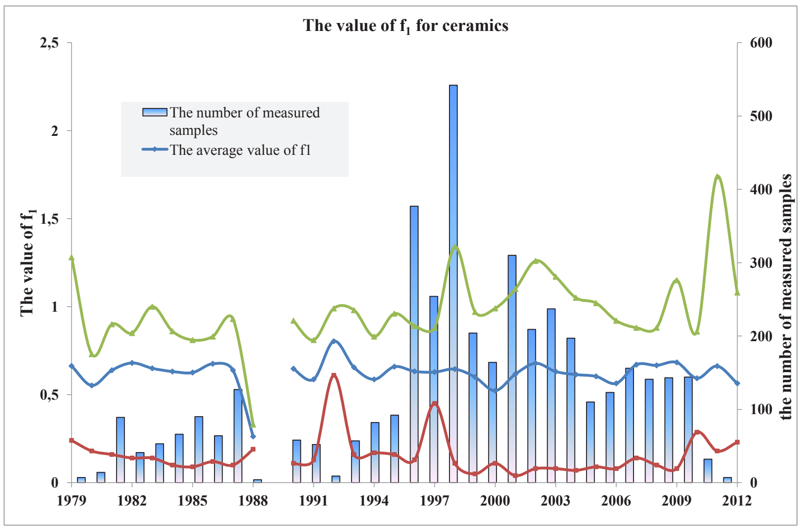

Fig. 9. The value of f_1 for ceramics at different times
Source: Own elaboration.

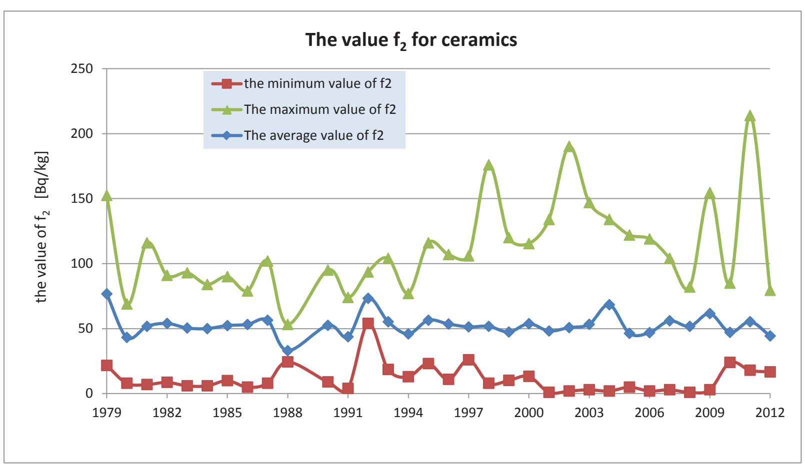

Fig. 10. The value of f_2 for ceramics at different times
Source: Own elaboration.

Average value of the activity coefficients f_1 for ash is above limit value f_{1lim} = 1.0, but below permissible value ($f_2 \leq 1.2$) if we want to use them for the construction of dwellings (except 2011). Average value of the activity coefficients f_1 and f_2 for ash ranges from 0.99 to 1.26 (Fig. 3) and 102.4Bq/kg to 135.1Bq/kg (Fig. 4) respectively [3]. Maximum value of f_2 was above the limit value for the construction of dwellings almost every year (Fig. 4). Although after 1991 the number of measurements of samples significantly increased, the average value of the activity coefficients f_1 and f_2 for ash remains at a relatively constant level, which can be caused by constant concentration of natural radionuclides in the ash.

Average values of the activity coefficients f_1 and f_2 for cement are well below limit values in the period of 1979-2012. The range of average value of the activity coefficients f_1 and f_2 for cement is from 0.26 to 0.60 for f_1 (Fig. 5) and from 34.3 Bq/kg to 82.6 Bq/kg for f_2 (Fig. 6).

Average values of the activity coefficients f_1 and f_2 for concrete are in the range from 0.52 to 0.81 for f_1 (Fig. 7) and from 50.3 Bq/kg to 84.8 Bq/kg for f_2 (Fig. 8) and ratio f_2 to f_1 remains almost equal 100.

Average value of the activity coefficients f_1 and f_2 for ceramics are in the range from 0.26 to 0.80 for f_1 (Fig. 9) and from 33.0 Bq/kg to 76.7 Bq/kg for f_2 (Fig. 10).

The table 1 shows the average value of the qualification coefficients f_1 and f_2 and a dose rate **for the selected materials.** The level of the qualification coefficients f_1 is **exceeded** only for the ash**, but the dose rate is still below 0.3 μSv/h.**

Table 1. Average dose rate with min and max value for selected materials in the period 1979-2012

Material	Average f_1 / Range of average f_1	Average f_2 / Range of average f_2 [Bq/kg]	Average dose rate / Range of average dose rate [nGy/h]
Carbon	0.24 (0.15 ÷ 0.43)	26.2 (14.7 ÷ 44.2)	31.8 (19.5 ÷ 56.2)
Ash	1.08 (0.99 ÷ 1.26)	119.2 (102.4 ÷ 135.1)	140.8 (128.0 ÷ 152.7)
Cement	0.38 (0.26 ÷ 0.60)	48.9 (34.3 ÷ 82.6)	50.5 (36.9 ÷ 78.5)
Concrete	0.66 (0.52 ÷ 0.81)	70.3 (50.3 ÷ 84.8)	86.5 (67.1 ÷ 105.5)
Ceramics	0.62 (0.26 ÷ 0.80)	52.6 (33.0 ÷ 76.7)	81.0 (34.2 ÷ 104.8)

Source: Own elaboration.

4. Conclusion

The qualification coefficients f_1 and f_2 are almost constant only for ash taking into account the entire period of the monitoring of the raw and building materials. It means is important to control all of them. The highest level of the qualification coefficients f_1 and f_2 are for ash and the lowest is for carbon – it is due to the process of the concentration of the radionuclides caused by the combustion process.

The concentration coefficient for the combustion is 4.5 on average. Higher values of qualification coefficients f_1 and f_2 characterize the materials containing considerable amounts of industrial raw materials such as ash or slag.

The level of qualification coefficients f_1 and f_2 for the ceramics is approximately twice as low as for the ash. The average values of f_1 during the study period since 1979 have not exceeded (except for ash in 2011) the limit for the applications in the construction of dwellings ($f_1 < 1.2$).

Although the average value of qualification coefficient f_2 for ceramics and cement is at similar level, the average value of f_1 for ceramics is almost 2 times higher, because concentration of ^{40}K dominates in natural activity.

The analysis of the number of values higher than the limits for the activity coefficients f_1 and f_2 for dwellings and public constructions in 1979-2012 shows that the trend of the average values of the activity coefficients f_1 and f_2 has remained constant for the last years.

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Literature

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