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The national survey of natural radioactivity in concrete produced in Israel

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A R T I C L E I N F O

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A B S T R A C T

The main goal of the current survey was to collect the results of the natural radiation tests of concrete produced in the country, to analyze the results statistically and make recommendations for further regulation on the national scale.

Totally 109 concrete mixes produced commercially during the years 2012–2014 by concrete plants in Israel were analyzed. The average concentrations of NORM did not exceed the values recognized in the EU and were close to the values obtained from the Mediterranean countries such as Greece, Spain and Italy.

It was also found that although the average value of the radon emanation coefficient of concrete containing coal fly ash (FA) was lower, than that of concrete mixes without FA, there was no significant difference between the indexes of both total radiation (addressing gamma radiation and radon together), and gamma radiation only, of the averages of the two sub-populations of concrete mixes: with and without FA.

1. Introduction

As known, most building materials of terrestrial origin contain small amounts of natural radionuclides. These radionuclides cause two types of radiation and radiation exposure: external and internal. The external radiation is caused by the gamma emitting radionuclides, which in the uranium series mainly belong to the decay chain segment starting with radium ^{226}Ra. The internal (inhalation) radiation exposure is due to radon ^{222}Rn (belongs to the ^{238}U decay chain), and marginally to its isotope thoron ^{220}Rn (belongs to the ^{232}Th decay chain), and their short-lived decay products, exhaled from building materials into the room air. Radon ^{222}Rn and thoron ^{220}Rn, gaseous radioactive progenies of ^{238}U and ^{232}Th, emanate from the soil and building materials, and along with its short-lived alpha-emitting descendants constitutes a source of internal exposure through inhalation. Radon is the most important source of exposure to natural radiation; it determines about 50% of the total dose from natural radiation (UNSCEAR 2008, 2010).

Knowledge of the concentration of natural radionuclides in building materials is important in the assessment of population exposures, as most individuals spend 80% of their time indoors and natural radioactivity in building materials is a source of indoor radiation exposure (Krisyuk, 1989; Zikovsky and Kennedy, 1992; Othman and Mahrouka, 1994). Indoor elevated external dose rates may arise from high activities of radionuclides in building materials.

The most important of natural radionuclides are ^{40}K and members of two natural radioactive series, which can be represented by the isotopes ^{226}Ra and ^{232}Th. The presence of these radioisotopes in the building materials causes external exposure to the people that live in the house. ^{226}Ra and ^{232}Th can also enhance the concentration of radon isotopes ^{222}Rn and ^{220}Rn and of its daughters in the house. ^{40}K and part of the decay series radionuclides mentioned before cause external exposure, while the inhalation of ^{222}Rn and ^{220}Rn and their short lived progeny lead to internal exposure of the respiratory tract to alpha particles (Keller et al., 1987; Savidou et al., 1996).

2. Limitation of the content of natural radionuclides in the norms

The convenient parameter used in the norms to limit the overall content of radionuclides in concrete and other building materials of mineral origin is so called activity concentration index. The activity index in the EU Directive (Council Directive 2013/59/Euroatom, 2014) and in many other national standards regulating radioactivity of

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building materials is calculated on the basis of the activity concentrations of radium (^{226}Ra) in the uranium (^{238}U) decay series, thorium (^{232}Th) in the thorium (^{232}Th) decay series, and potassium (^{40}K). Other nuclides are sometimes taken into consideration as well: for example, the activity concentration of cesium (^{137}Cs) from fallout is regulated in the Finnish guidelines (ST12.2, G, 2005).

Usually, if the activity index exceeds 1, the responsible party is required to show specifically that the relevant action level is not exceeded. If the activity index does not exceed 1, the material can be used, so far as the radioactivity is concerned, without restriction.

The criterion of meeting the standard is the non-dimensional value of so called activity concentration index taking into account the total effect of three main natural radionuclides, which can present in building materials:

$$I = \frac{A_{Ra}}{300} + \frac{A_{Th}}{200} + \frac{A_K}{3,000} \leq 1 \quad (1)$$

where A_{Ra}, A_{Th} and A_K are activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, respectively, in Bq kg^{-1} (Council Directive 2013/59/Euroatom, 2014).

The Member States are required to insert in their legislation a list of the different types of building materials which need to be controlled with regard to their emitted gamma radiation. Materials incorporating by-products or residues from NORM industries, such as fly ash, are in this list.

In spite of the fact that the overall radiation hazard due to ionizing radiation from building materials includes both gamma radiation component, which depends on their radionuclides content, and the component caused by their radon exhalation, most of the standards in the world, which regulate radioactivity of building materials, concrete including, address the gamma radiation only (Kovler, 2011). Indeed, the evaluation of the excess dose caused by building materials for the radon pathway is rather complicated. One of the reasons is that the actual correlation between the radon exhalation rate measured in laboratory and the excess indoor radon concentration on site might be rather poor (Markkanen, 2001). Numerous factors, such as temperature (both indoors and outdoors), air pressure and humidity fluctuations, total porosity, pore distribution and pore type (open or close), surface treating done at the building site or type of the coating material applied, influence significantly radon exhalation in dwellings. Finally, it is extremely difficult to take into account the effect of the inhabitant behavior – first of all concerning ventilating. That is why most of the standards regulating radioactivity of building materials address the radon exhalation in a very simplified form – through the limitation of ^{226}Ra – the parent of ^{222}Rn in the ^{238}U radioactivity chain.

3. Concrete as a source of natural radiation indoors

Concrete is the most popular building material in the world: annual production of concrete is about 1 m^3 per capita (in Israel it is even higher, about 2.5 m^3 per capita).

The results obtained in Finland, Greece, Germany and the Netherlands (RP-96, 1997) show that concrete has the highest normalized radon exhalation rate among building materials (exhalation rate per unit area and per unit content of ^{226}Ra), about 0.2–0.5 (Bq m^{-2} h^{-1})/(Bq kg^{-1}). Therefore, concrete in buildings can contribute to indoor radon levels more than other building materials with the same ^{226}Ra content. This fact can be explained by relatively thick building elements (walls, floors and ceilings) made of concrete, and high specific surface area of cement hydrates, which makes easier release of radon atoms into the porous microstructure and their transport to the surface (Kovler, 2012b).

Concrete is widely used for dwelling construction in Israel. Dwellings in Israel must include a Residential Protected Room (RPR) made of thick concrete slabs and walls, with an extremely airtight window and outward opening extremely airtight steel door; while an inward opening regular door is applied for everyday usage (Becker et al., 2013). Being multi-purpose, the RPR raised concern regarding long-term radon exposure.

4. Controversial radiological effects of using coal fly ash in concrete

The recycling of coal fly ash (FA) (in particular, in concrete construction) has become a challenge in recent years due to increasing landfill costs and current interest in sustainable development. Some concrete plants in Israel produce concrete with FA as a mineral additive replacing partially Portland cement (because of pozzolanic properties of FA contributing in strength and durability of concrete). However, FA can successfully replace also the fine fraction of sand, which becomes especially important nowadays because of the lack of high-quality quartz sand from dunes located along the Mediterranean seashore, from where its extraction is banned by the environmental authorities. In addition, FA as a replacement of fine sand improves workability and pumpability of fresh concrete mixes. As a partial replacement of sand, fly ash can be introduced in normal-weight concrete mixes by much larger amounts, than replacement of cement. At the same time, the level of 120 kg m^{-3} is usually not exceeded in concrete mixes applied in construction of habitable structures in Israel.

The use of FA in concrete and other building materials is a well-recognized source of gamma exposure that is due to the presence of activity concentrations of the three primordial radionuclides, ^{226}Ra, ^{232}Th and, to a lesser extent, ^{40}K, while the effect of coal fly ash via radon exhalation is controversial, in particular, due to the low emanation coefficient of the ash particles (Kovler et al., 2004).

5. Motivation and goals of the national survey

The national surveys of ionizing radiation characteristics in concrete are an essential basis for regulation aimed to protect the population from the exposure to natural ionizing radiation in residential buildings.

To date, these requirements in Israel were based on the limited data about the current situation in ionizing radiation of construction products.

The research team led by the author was requested by the Israeli Ministry of Environmental Protection to conduct the national survey of natural radioactivity of representative concrete mixes produced commercially by the local concrete plants. The main goal of the survey was to collect the results of the natural radiation tests of concrete produced in the country, to analyze statistically the test results of routine inspections conducted by the commercial laboratories working according to the standard SI 5089, and to make recommendations for further regulation on the national scale. The survey was aimed to check whether the assumptions behind the calculations used as a basis in the standard requirements, are close to the reality.

The main goal of the current survey was to collect the results of the natural radiation tests of concrete produced in the country, analyze the results statistically and make recommendations for further regulation on the national scale.

6. Regulation in Israel

The tests were conducted during the years 2012–2014, according to the Israeli Standard (SI 5098, 2009). The purpose of the Standard is to allow systematic control by the authorities on the concentration of radioactive elements from natural sources in