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RADIOECOLOGICAL STUDIES IN ARMENIA: A REVIEW

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ABSTRACT

This review generalizes results of radioecological studies implemented in Armenia in a period from the 1960s till the early 2000s. Specificity of geographical position determines the maximum of global atmospheric radioactive fallout in high mountain regions of the country. Several facilities can potentially influence the radioecological situation of Armenia: the Armenian Nuclear Power Plant, a spent nuclear fuel repository and interim storage facility for radioactive waste. Runoff of many rivers of the basin of Araks and Kura (major water arteries of South Caucasus) originates on the territory of Armenia. The aforesaid peculiarities determine the key role of Armenia in transfer and distribution of radionuclides in the region.

Key words: *Environmental Radioactivity; ^{137}Cs; Soil; Bottom Sediments; Building Materials*

INTRODUCTION

Due to geographical position, complex relief, presence of a nuclear power complex and other specificities, the territory of the Republic of Armenia plays a crucial role in transboundary transfer and redistribution of radionuclides in the South Caucasian region [3, 17, 21, 23, 27].

The role Armenia has in transboundary transfer of radionuclides in the South Caucasian region includes a few underlying factors such as global radioactive fallout [39], radionuclide accumulation in depositing environments throughout Armenia's highlands, and transboundary transport of radionuclides with river runoff [2, 3, 17, 21, 23, 25, 27]. Hypsometrically, as compared with the rest of the South Caucasian republics, Armenia is positioned at the highest height, mean 1830 m a.s.l. The runoff from plenty of tributaries of Rivers Kura and Araks, the largest water arteries of the South Caucasus, initially originates on her territory [42].

Besides transboundary pollution, there exist a number of potential sources of radionuclides throughout Armenia, and in the neighboring countries. Armenia houses an operating NPP, a spent nuclear fuel repository and a regional low and intermediate level waste repositories [10, 16, 30, 31]. Armenia borders with the countries which actively develop nuclear technologies, atomic energy and uranium mining as well [19].

Thus, radioecological research in Armenia is both scientific interest and security importance. This review article generalizes results of implemented radioecological studies in order to characterize the distribution of radionuclides in Armenia, evaluation of background and baseline levels of naturally occurring radioactive materials (NORM) in different environmental compartments, as well as to assess cross-border radioactive pollution. Different aspects of environmental impact of ANPP are considered in detail and introduces in the number of publications [10, 12, 13, 16, 31] and therefore, this topic will be omitted in the current review. The review covers the radioecological and investigations conducted in Armenia from 1950s to 2010s.

GEOGRAPHY AND CLIMATE OF ARMENIA

Armenia is located between 38°50'-41°18' northern latitude and 43°27'-46°37' east longitude, in the northwestern part of the Armenian Highland, occupying most of the interfluve between the Kura and Araks rivers and borders by Georgia, Iran, Turkey, Republic of Artsakh and Azerbaijan. The relief is predominantly mountainous [42].

Mountain range of the Lesser Caucasus stretches to the southeast, across the northern part of Armenia, where the relief is composed of medium-altitude mountain ranges separated by deep valleys. The Eastern Armenian volcanic range, occupying about a third of the country's territory, runs from the mountain range of the Caucasus to southwest. Those volcanic mountain chains extend from the Javakheti range in the northwest to the Garabagh range in the southeast. Here, the relief is rugged, the main forms of relief are lava plateau, erosion valleys, volcanic ridges (Gegham, Vardenis) and massifs, and the largest is Aragats. The southern part of the country is an area with blocky mountains and deep river valleys. Relief features of this area include high ridge height (Zangezur Ridge), deep and dense relief separation and well-pronounced altitudinal zonality [42, 43].

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The climate of Armenia is diverse. The temperature depends mainly on the altitude. Despite the subtropical zone location, the subtropical climate is detected only in the southern part of Armenia (Meghri). In other areas, the climate is alpine, continental with hot summers and cold winters. On the plains, the average January temperature is –5°C, July +25°C; in the mountains –6°C and +20°C, respectively. The minimum amount of precipitation in Ararat valley (850-1000 m a.s.l.) is 200-250 mm per year; in the middle mountains (from 1700 to 2500 mm a.s.l.) – 500 mm; in highlands (more than 2500 m a.s.l.) – 700-900 mm [14].

INITIAL RADIOECOLOGICAL STUDIES

The first radioecological studies in Armenia were carried out in 1958 at the Institute of Agrochemical Problems and Hydroponics (IAPG) of the Academy of Sciences of the Armenian SSR. These studies were aimed at assessing gross radioactivity of soils and are valuable as initial stage of data collection [18].
In the subsequent works of the IAPG in the 1970s was estimated the distribution of naturally occurring radionuclides in soils (^{238}U, ^{226}Ra, ^{232}Th, ^{40}K, ^{87}Rb). It was found that the content of ^{238}U, ^{226}Ra and ^{232}Th in soils conditioned by geological structure of the area [5]. With high levels of ^{238}U stand out chernozem and mountain-meadow soils of Armenia [29]. Activity concentrations of ^{40}K and ^{87}Rb in soils depended on the type of soils, in particular, the presence of clay fraction and the amount of organic matter [5, 11]. Although these results are not comparable with later data, they are valuable for characterization of natural radionuclides distribution in different soil-climatic zones of Armenia.

CROSS-BORDER TRANSFER OF RADIONUCLIDES

The radioecological situation in certain territories is conditioned not only by local characteristics but also by the transfer of radioactive substances from neighboring territories. Nuclear weapon tests, accidents in NPP and releases from nuclear industrial complexes led to the global spreading of radionuclides [40].
The maximum level of gross β-activity of atmospheric dust in Armenia was reported in 1970 and 1971 and was decreased to 4 times in 1983. After the Nuclear Test Ban Treaty occurred in 1963, the radioactivity of atmospheric deposition is mainly represented by long-lived ^{137}Cs and ^{90}Sr. The distribution of the concentration of these radionuclides in atmospheric precipitations and fallout occurs along high altitudinal zones [1, 3]. ^{137}Cs and ^{90}Sr associated with the airborne dust after precipitation accumulated mainly in upper soil layers. The distribution of ^{137}Cs and ^{90}Sr by profile differs depending on soil type [4, 6].
The largest nuclear accident was the Chernobyl NPP in 1986. According to a number of studies, the territory of Armenia was also influenced by radioactive contamination. In the soils of the western slopes of Geghama Range (2500 m above sea level) and Aragats massif (3250 m above sea level), in 1986 a short-lived ^{134}Cs was detected, which was not detected in the lowlands (850-1000 m). After 1986, the spread of concentrations of long-lived ^{137}Cs and ^{90}Sr in soils indicate a "fresh" input of these radionuclides brought with air streams [2, 3, 7, 16, 42]. Similar results were obtained in the Ararat valley. In 1986 the concentrations of ^{137}Cs and ^{90}Sr in air and atmospheric dry depositions, compared to the subsequent period (***Fig. 1***), increased more than two orders of magnitude. However, since 1987, activity concentrations of

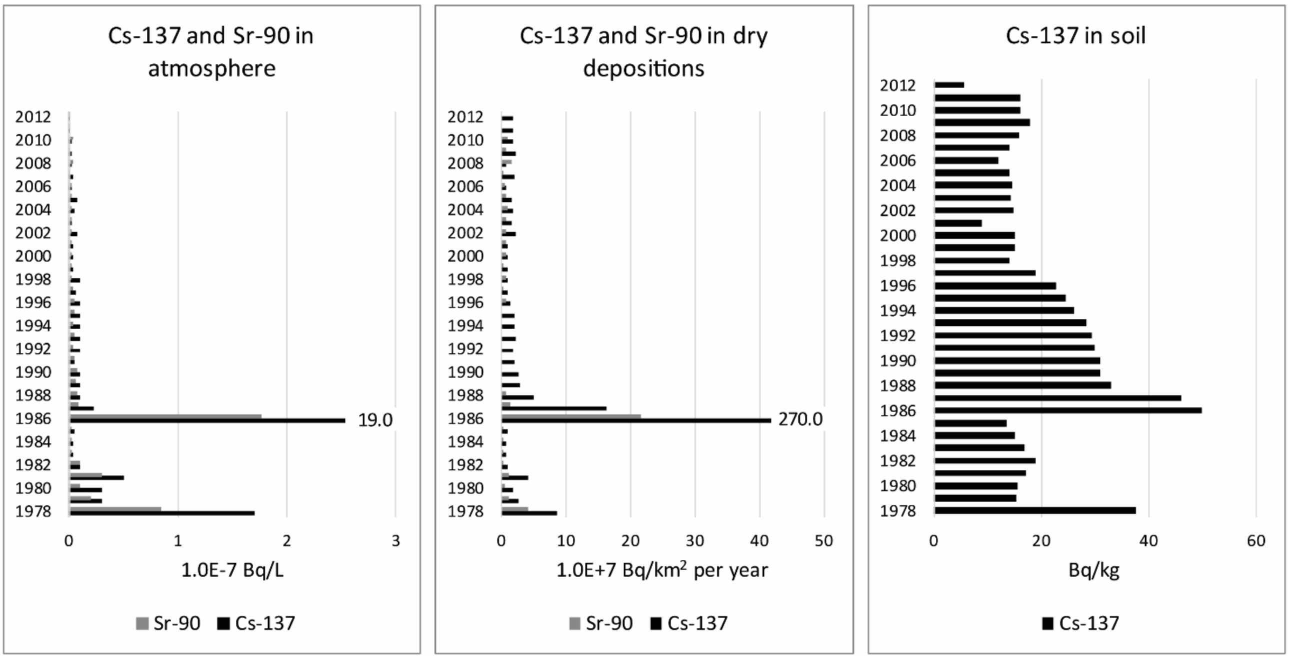

Fig. 1. Average mid-annual concentrations of ^{137}Cs and ^{90}Sr in atmosphere (1.0E-7 Bq/L) and dry atmospheric depositions (1.0E+7 Bq/km^2) (adopted from [13, 30, 31]) and the average concentrations of ^{137}Cs in soils (Bq/kg) of Ararat valley (adopted from [13])

^{137}Cs and ^{90}Sr have returned to "pre-Chernobyl" values. Several years after the accident, the content of ^{137}Cs in soils

(***Fig. 1***) was remained relatively high and reached pre-Chernobyl values up to the 2000s [30, 31]. In addition, decay products of ^{235}U were detected in the atmospheric air in May of 1986 (***Tab. 1***). In May and June ^{131}I and ^{137}Cs (***Tab. 2***) were identified in cow milk samples as well [30].

Table 1. *Concentrations of several radionuclides in atmospheric air (1E-7 Bq/L) in Ararat Valley in May-June 1986 [13]*

Radionuclide	^{144}Ce	^{141}Ce	^{103}Ru	^{106}Ru	^{95}Nb	^{95}Zr
Activity concentration, 1E-7 Bq/L	14.97	2.83	47.7	335.5	1.0	1.3

Table 2. *^{131}I and ^{137}Cs concentrations in cow milk samples (Bq/L) [16]*

Radionuclide	Sampling date			
	20 May 1986	29 May 1986	12 June1986	26 August 1986
^{131}I	27.3	4.8	4.8	Not detected
^{137}Cs	49	3.7	8.1	1.3

‘‘South Caucasus River Monitoring’’ NATO Science for Piece № 977991 project (CENS, NAS RA, 2003-2008) results revealed low radioactivity in the river of Armenia. Natural radionuclides ^{40}K and ^{226}Ra were found in the waters (***Fig.2***). From 2006 to 2008, the activity concentration of ^{40}K in various rivers ranges from 0.10 to 1.91, with an average

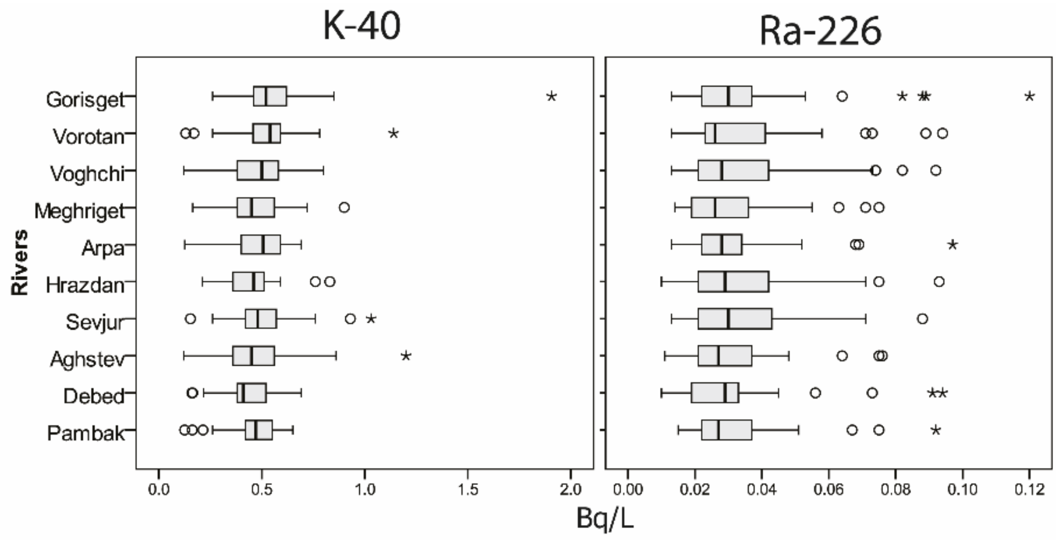

Fig. 2. Distribution of ^{40}K and ^{226}Ra concentrations in the largest rivers of Armenia in the period of 2006-2008 (modified from [32])

of 0.35 Bq/L; ^{226}Ra - from 0.01 to 0.12, with an average of 0.03 Bq/L. During the entire study period, ^{137}Cs was fixed in five samples of river water: from 0.001 to 0.004 Bq/L [17, 27, 32].
The results of studies substantiated the approaches for the creation of Rapid Response systems within the frames of radionuclides cross-border transfer control in the South Caucasus region [27].

PALEOLIMNOLOGY AND PALEO-RADIOECOLOGY OF LAKE SEVAN

Paleo-limnological and paleo-ecological studies of Lake Sevan were implemented in CENS in cooperation with Department of Oceanography of Florida State University (FSU) and Department of Oceanography and Coastal Sciences of Louisiana State University (LSU) in 2001-2003 [8, 22]. The mentioned particular research was aimed at revealing the natural variations that occurred prior to the 1930s as and subsequent radioactive global fallout.
Seven bottom sediment core samples were collected from Lake Sevan in September 2002. Activity concentrations of ^{226}Ra, ^{137}Cs, ^{40}K, ^{232}Th, ^{210}Pb in samples were determined using HpGe well-type detector. ^{210}Pb dating was performed via CRS and CIC models [9]. ^{210}Pb was concentrated mainly in upper layers (0-30 cm). Deeper, ^{210}Pb approaches equilibrium with ^{226}Ra. Excess ^{210}Pb originates from atmospheric decay of ^{222}Rn. ^{226}Ra was deposited in lake sediments with erosional inputs. As seen, beginning from some 30 cm ^{210}Pb activity in sediments was changed in parallel to ^{226}Ra. ^{137}Cs accumulated in the upper sediment layer (0-30 cm); occasionally showing a double-maximum probably related to global fallout (1962-63) and the Chernobyl accident (***Fig. 3***). The method of ^{210}Pb dating enables to calculate sedimentation rates and evaluate environmental changes induced by eutrophication, erosion-siltation, acid deposition, and other human disturbances of the Sevan basin.

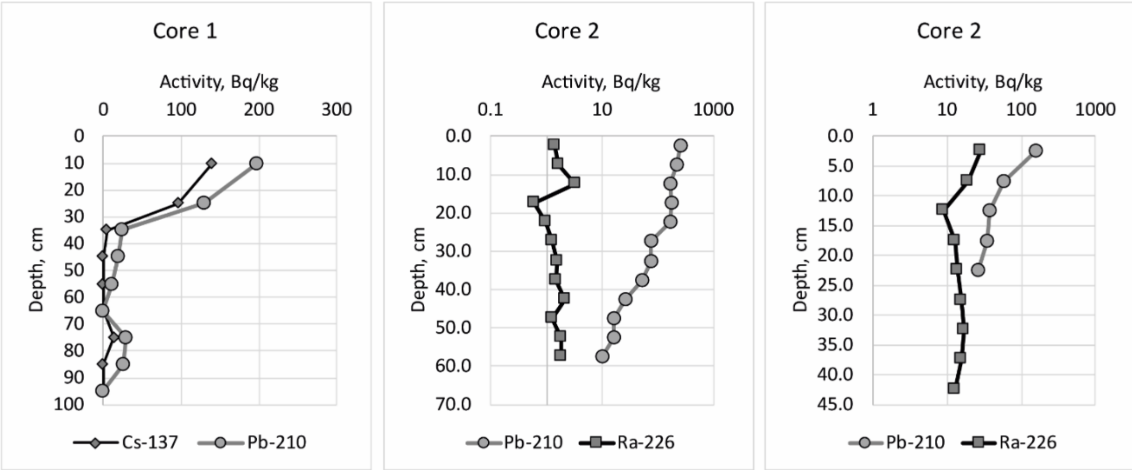

Fig. 3. *^{137}Cs, ^{210}Pb and ^{226}Ra activity concentrations in Small Sevan bottom sediments (modified from [21, 22])*

URBAN RADIOECOLOGICAL STUDIES

Urban radioecological studies in Armenian were carried out since 1958. Studies covered different medium of urban environment: soil, atmospheric deposition, etc. According to obtained results the levels of gross beta activity of dry atmospheric depositions in Yerevan during 1971-74 varied 111.0-1857.4 with an average 672 Bq/m^2 per year [1, 3, 7, 24, 26].

After the Chernobyl NPP accident in 1986, high activity concentrations of artificial radionuclides ^{137}Cs and ^{90}Sr were recorded in the atmospheric depositions of Yerevan. In 1989-2001, the gross beta activity of dry atmospheric depositions decreased up to 304 Bq/m^2 per year. Concentrations of ^{137}Cs and ^{90}Sr decreased as well, in 2 and 3 times respectively [3, 7, 25].

More informative data was obtained as a result of a comprehensive urban soil survey (Sc. 1: 50000) implemented in Yerevan in 1990 and 2002. The first soil survey of 1990 reflects the radioecological situation of Yerevan, which was formed after nuclear weapon testing in Northern Hemisphere and the Chernobyl accident. In 1990 the soils of Yerevan were characterized by the high variability of gross beta-activity (V = 6.5). The values range from 538 to 916 Bq/kg, the high values (> 800 Bq/kg) recorded in 11% of soil samples. The distribution of gross beta-activity was mosaic (***Fig. 4***, ***a***) [7, 26].

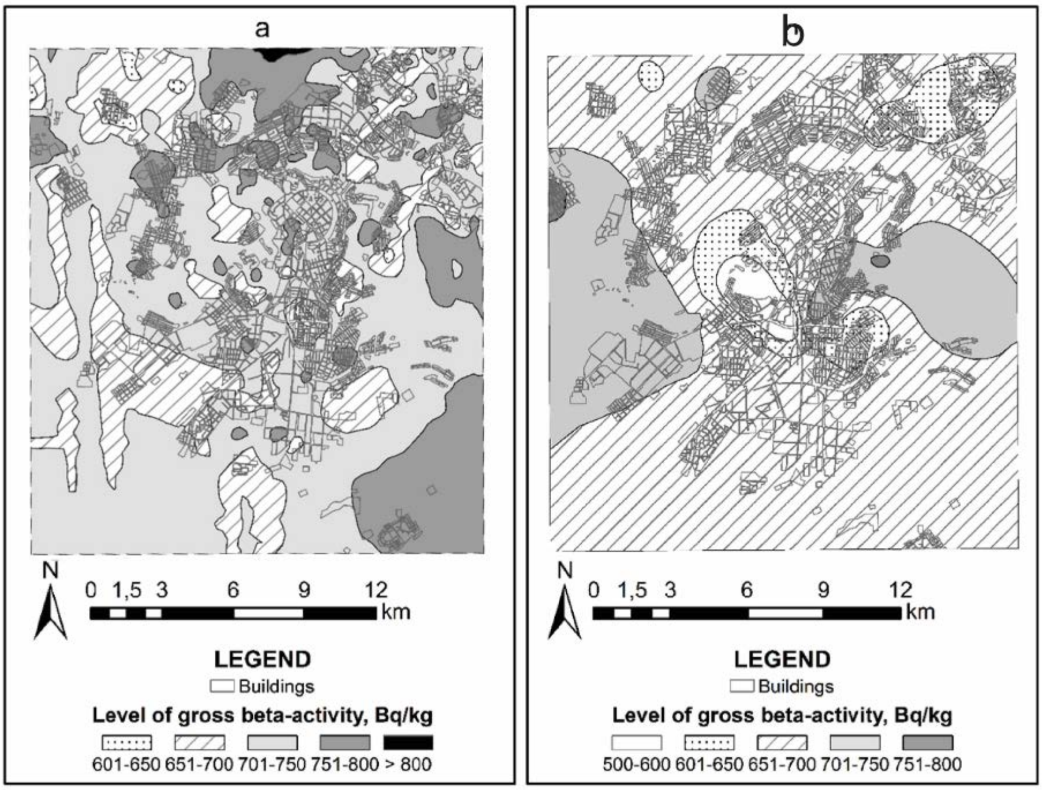

Fig. 4. *The spatial dynamics of gross beta activity in the soils of Yerevan in 1990 (a) and 2002 (b) (modified from [7, 26])*

In the period of 1990-2001, gross beta-activity decreased in atmospheric depositions after nuclear weapon tests ban. Subsequently, beta-activity of urban soils decreased as well (***Fig 4, b***) and the distribution of beta-activity levels was more homogeneous [24, 26].
In 2005 a similar soil survey was carried out in the town of Kajaran, which is one of the largest mining centers of Armenia. The studies in the 1950s showed the presence of 3 uranium mineralization zones in Kajaran [38], however, the possible radiological hazard to population had never studied there. According to pilot radiological investigation results the average value of gross beta-activity in the soils was 736 Bq/kg, which exceeds the estimated mean level: 500-600 Bq/kg. The level of beta activity is gradually changing in Kajaran, where the highest levels are attached to the production complexes, including open-pit mining and copper-molybdenum combine. The residential part of the city is mainly characterized by the average level of gross beta activity (758-808 Bq/kg) [38].
The urban radioecological studies were performed using different analytical equipment with different sensitivity, accuracy, and resolution which makes the results incomparable. Therefore, data should be revised in order to assure the continuity of assessment of urban environmental radioactivity dynamics.

RADIOACTIVITY OF CONSTRUCTION AND BUILDING MATERIALS

In 1983-85 concentrations of NORM in the Armenian construction and building materials were investigated by a group of hygienist from Research Institute of General Hygiene and Occupational Diseases after. N.B. Hakobyan of the Ministry of Healthcare of ArmSSR [28]. The main tasks of that study were to determine the content of NORM (^{226}Ra, ^{232}Th, ^{40}K) in local construction and building materials and to assess exposure to the population.
In 1983, 28 state companies were operating in the republic that produced 20 main types of stones and by-products. Studies covered all exploited deposits and types of produced building materials in the republic.
Radioactivity of building materials was studied using low-background “УМФ-3” with “СИ-16 БГ” detector and “УМФ-1500М” end-counter “СБТ-13” for gross beta and gamma activity measurements, respectively. Isotopes were identified by using scintillation gamma spectrometer СГС-200. High values of specific radioactivity were recorded for tuffs, pumice, volcanic slag and granites, the lower levels – for limestone and marble. The maximum values of the average specific gamma activity recorded in pumice blocks: 75.436±0.456 pCi/g. Gross beta-activity was higher in tuff and partition blocks: 43.112±1.732 and 43.055±1.68 pCi/g, respectively [28].
The average effective specific activity from NORM in construction and building materials was 3.811 pCi/g, which exceeded the average value: 2.70 pCi/g, accepted in the Soviet Union.
The greatest variability in the concentrations of natural radionuclides is characterized by tuff, basalt and felsite (***Fig. 5***) since the activity concentrations of radioisotopes strongly depend on the conditions for the formation of deposits. The activity concentrations of radionuclides in the studied building materials did not exceed the standards of radiation safety accepted for that particular period and consequently could be used for any types of construction without limitations.

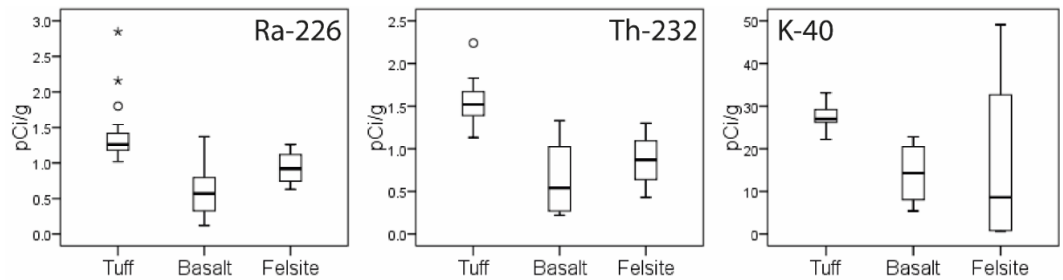

Fig. 5. *Boxplots of the activity concentrations of NORM in natural constructing material (modified from [28])*

The annual effective dose equivalent of NORM from building materials was 632 μSv annually, which twice exceeds the international norm: 350 μSv [41] and the average value of external gamma-irradiation (370 μSv), including background and additional gamma dose from building materials, was also assessed for the population of the USSR [20].
The radioactivity of natural building materials remains the issue to be addressed. Further investigations should conduct taking into account local use of building stones and by-products and their export rates as well.

INDOOR RADON AS AN ENVIRONMENTAL CONSEQUENCE OF EARTHQUAKES

Indoor radon monitoring and assessment of radon-induced health risk to humans is a pressing issue for cities with active geodynamic processes determined by the presence of active geological faults, epicenters of earthquakes, etc.
Yerevan characterizes by active geological faults, with the risk of radon-induced adverse health effects in humans being rather high. The researches on indoor radon concentration dynamics have been implemented in Armenia since early 1970s [33, 34], but the most comprehensive data were collected in CENS between 2005 and 2010 [15, 35, 36, 37]. The studies were aimed at a comparative analysis of the concentration of indoor radon depending on geodynamic processes – earthquakes associated with fault zones found throughout Yerevan. Radon concentration measurements were done using a portable radon detector RAD 7 (DURRIDGE, USA) in the basement. To obtain mean daily concentration, 3 measurements were done daily and the results averaged. Data on radon concentration were collated with those from

earthquake catalogs provided by National Survey for Seismic Protection of Armenia; Kandilli Observatory and Earthquake Research Institute, Bogazici University, Turkey; Iranian Seismological Center of the University of Tehran, Iran.

While analyzing available data, there were selected the earthquake epicenters located only within a 50 km radius around Yerevan or lying on faults associated with its area. Data collation has indicated that each earthquake is preceded by a peak value of radon concentration. On geodynamical active days, radon concentration in the air of the basement space was relatively decreasing but increased after activity decay. Maximal concentrations were detected after a series of small earthquakes that occurred between December 2006 and March 2007 [15, 37]. The average radon concentration in the air was 908.8 Bq/m^3, it's three times higher than a reference level for indoor radon recommended by WHO.

Further monitoring studies are required in order to detect the precise dependence of indoor radon concentration dynamics and determine an effective dose for the population as a consequence of geodynamic activity of the region.

CONCLUSION

This review summarizes radiecological investigations implemented in Armenia from from 1950s to 2010s. The initial studies were implemented in order to assess the activity of naturally occurring radionuclides in different landscapes and reveal correlations between radioactivity, soil properties and type of land use. This valuable information enables the creation of a national inventory of naturally occurring radioactivity however the results obtained before the 2000s are not comparable with later data. Therefore continuity of radioecological data should be one of the main challenges to be addressed in future. The radiological survey of urban areas revealed the importance of monitoring naturally occurring radionuclides and related radon hazard assessment. Further work is needed to assess the share of different sources in technologically enhanced natural radioactivity.

Abundant data characterizing the alteration of radiological background after nuclear weapon testing and the Chernobyl NPP accident have been generated in Armenia. Analysis of Chernobyl accident consequences for Armenia reported by various research teams in different years enables revealing the main pathways of transboundary radioactive pollution and redistribution of radionuclides throughout Armenia with the relevant assessment of potential risk to public health. Distribution and migration of artificial radionuclides in the environment should be studied further in order to complement the data essential for early warning and rapid response system within the frames of national radiation protection and nuclear security.

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