Polish Journal of Environmental Studies Vol. 6, No. 6 (1997), 51 - 55

Radioecological Analysis of the North-Eastern Region of Poland

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Received 30 March, 1997
Accepted 15 July, 1997

Abstract

This paper presents the results of measurements of natural radioactivity of K-40, Ra-226 and Th-232 in building material as well as radon concentrations and dose rates in houses and outdoors. The mean absorbed rate in air outdoors from natural gamma radiation was found to be 40.2 $nGyh^{-1}$. Doses inside houses made from particular materials were ($nGyh^{-1}$): brick - 102, prefabricate - 89, wood - 88. The mean equivalent concentration of radon indoors was 12 Bqm^{-3}. The calculated annual effective dose equivalent per one statistical inhabitant of "Poland Green Lungs" is 1.61 mSv/year. The doses absorbed by the population in the northeastern region of Poland from natural sources of ionizing radiation are lower than the population-weighted world averages.

Keywords: natural radioactivity, radioactivity in building materials, radioecology

Introduction

The northeastern area of Poland is a region featuring high tourist qualities with low standards of urbanization and industrialization. In 1990, a special functional area was cut out and called ''Poland's Green Lungs''. This area, approx. 60 thousand km^2 (19.4% of the total area of the country), is at present inhabited by about 3.7 million people and includes the following provinces (called voivodships): Białystok, Łomża, Ciechanów, Olsztyn, Ostrołęka, Suwałki and parts of Elbląg and Siedlce [1]. In view of the present situation of the natural environment, this region has been incorporated into the system of ecological development [2] and, in the future, may become a frame of reference for the evaluation of the effects of civilization processes. This is why this area should become a specimen for environmental analysis, including analysis of the radioecological situation. This analysis has been made possible studies of natural radioactivity carried out for many years in the northeastern territory of Poland, including the provinces of Białystok, Łomża, Suwałki, Olsztyn and Ostrołęka, by the Department of Biophysics, Medical Academy in Białystok.

The average contributions to the exposed dose from the natural gamma radiation background are 35%, 25% and 40% for K-40, U-238 series and Th-series, respectively [3]. Thus the abundance of these radionuclides in soil and building materials are decisive as far as the doses absorbed by the population indoors and outdoors are concerned. These doses can be increased as a result of the collection of wastes from coal-burning power stations, mines or other steel and chemical plants. They all produce garbage and refuse such as dumps, sewage or sediment ponds or floatation waste tanks, which lead to higher environmental radiation levels [4-7]. The elevation of the background doses may also be caused by intensive fertilization by potassium and phosphorous compounds [8]. There is also growing concern that the background may increase as a result of introducing new building technologies based on by-products of the power, steel and chemical industries such as fly-ash, blast furnace slag or phosphogypsum [9-10]. Economically, these by-products can undoubtedly be of great value, but, radiologically, they may lead to a greater hazard [11].

The greatest contribution to the population hazard, up to 50% of the dose equivalent from naturally occurring sources, is obtained from the radioactive isotope of radon (Rn-222) and its decay products [12]. Thus, radon concentrations inside buildings depend mainly on the type of geological subsoil and the transfer of Rn-222 from soil air to the interior spaces in buildings, and also on the inherent content of Ra-226 in building materials [13-14].

This work presents results of measurements of K-40, Ra-226 and Th-232 concentrations in the subsoil and raw and building materials used in the construction of houses for permanent residence in northeastern region of Poland [15-16]. Results are also shown of direct measurements of gamma radiation background dose rates inside typical dwelling houses and in the open air [17-18], as well as of Rn-222 and its daughter product concentrations in the air inside houses [19-20]. These studies have made it possible not only to provide a pattern of natural radioactivity in the

region, but also to make appropriate comparisons with other results obtained in Poland and in the world, and to assess the population hazard of that region to radon and environmental gamma background.

Measuring Techniques

In the measurements the following techniques were used:

1. The measurement of the dose rate inside buildings and in the open air were performed using a high pressure ionizing chamber (current type) constructed by CLOR in Warsaw, and an electrometer with a dynamic capacitor (VA-J-51). The set calibration was made using Ra-226 source (3.7 GBq) taking into consideration the effect of scattering radiation and directional correlation of chamber sensitivity, and absorption in the air due to distance. This type of chamber and measuring method have been described in detail by Pełnsko [21]. The uncertainity of measurements using these methods depends on the precision of measuring the activity of the calibration source (about 10%). The measurements of the ionization chamber yield were made at Sawinda Lake in the province of Suwałki, at latitude 53° 32'.6 N and longitude 22° 13'.6 E, at 124.6 m above sea level.

2. Natural radioactivity measurements of raw and building materials and soil were made using a three-channel amplitude AZAR analyzer with a NaI detector. The technique used was that developed at the Institute of Construction Technology (Instytut Techniki Budowlanej) for all laboratories in Poland.

3. Measurements of radon and its products in houses and in their environment were made using two methods. For radon studies, the Markov technique was employed, in which the total activity of the radon alpha decay products collected on a filter (through which a certain volume of air was passed) was assayed [22]. In field studies, a mobile detection unit was used, consisting of an alpha radiation detector and a rotemeter. Using this method the measuring cycle was based on air filtering through a filter (mesh diameter 4 μm) during 5 min; the average air flow during 5 min was 57 dm^3 (ranging from 44 to 69 dm^3). The aerosol containing radioactive decay products of Rn-222 are deposited on the filter with efficiency $\eta = 0.8$. Next, two independent measurements of the radioactivity of the aerosol collected on the filter were performed in two different time intervals (7-10 s and 10-15 s). The efficiency of registration of alpha radiation at the used spectrometer set was $\varepsilon = 0.28$. The background counting with a clean filter was 15 imp/3 min. Independent series of radon studies were carried out in houses of Białystok province and in the open air using detectors with activated carbon. This method, described by A.C. George in 1984 [23], is recommended by the American Environmental Protection Agency (EPA) for environmental investigations of radon emanations. In these studies, passive PICO-RAD (Packard) detectors were employed, enabling mean radon concentrations in air to be determined during a two-day exposition. The readings were taken at the laboratory of the Department of Biophysics, Medical Academy in Białystok using a TRI-CARB liquid scintillation set. An Insta Fluor liquid scintilator (Packard) was used.

Results of Natural Radioactivity Measurements in the Environment

Dose Rates in the Open Air (Outdoors)

On the basis of measurements at 231 places in northeastern Poland, the mean absorbed dose rate in air from natural gamma radiation was found to be 40.2 $nGyh^{-1}$ (S.D. = 8.8 $nGyh^{-1}$) with the variability range of 6.1-84.2 $nGyh^{-1}$ [17]. The recent data included in the UN Report [12], representative of 3/5 of the population, provide the national mean value ranging from 24 to 160 $nGyh^{-1}$, and the weighted mean for the world's population of 57 $nGyh^{-1}$; this is higher than the value found for northeastern Poland. A similar regularity was confirmed by spectrometric measurements of 52 specimens of the topsoil collected in the same region of Poland [16]. The mean concentrations of the main gamma emitters and their variability ranges, in Bq/kg, were: 433(203-662) for K-40, 11(1-26) for Ra-226 and 15(6-26) for Th-232. These values are lower than those of the world's mean concentrations. The concentrations of radioactive potassium, radium and thorium in soil measured in northeastern Poland made it possible to estimate the dose rate from natural gamma radiation under the assumption that the distribution of these radionuclides is uniform and has a 2π geometry. The following calculation coefficients were used for the absorbed dose rate in air, corresponding to the activity of 1 Bq/kg: 0.0414 nGy/h for K-40; 0.461 nGy/h for Ra-226 and 0.623nGy/h for Th-232 [12]. The estimated dose rate for northeastern Poland was 32.3 nGy/h, lower than the value of 40.2 nGy/h resulting from the direct measurements of dose rates in the field, which accounted for the effect of other natural gamma emitters and radioactive soil contamination. From our studies it was found that about 55% of the exposition in the above region is due to K-40, 29% to Th-232 and 16% to Ra-226.

Dose Rates in Houses

The measurements made in 342 houses in northeastern Poland showed that the highest mean dose rates were observed in brick houses (102 $nGyh^{-1}$), and significantly lower values were found in prefabricated (89 $nGyh^{-1}$) and wooden (88 $nGyh^{-1}$) houses [18]. The national mean values quoted in the UN Report [12] based in most cases on model calculations using data on radionuclide abundances in building materials, range from 20 to 190 $nGyh^{-1}$, resulting in a mean dose rate of 80 $nGyh^{-1}$ for the world's population. Our measurements inside houses were carried out with the use of an ionization chamber and, therefore, they take account of the total dose from gamma emitters in building materials as well as the dose from the terrestrial and cosmic background (less the shielding effect of walls and ceilings).

Radon Concentrations

The most extensive measurements of radon products made in northeastern Poland have been reported in [19]. Some 185 buildings situated in the Białystok, Łomża and Suwałki provinces were investigated. The mean concentrations of Po-218, Pb-214, Bi-214 were found to be 15, 12

and 11 Bq/m^3, respectively. As a result, the mean potential energy of alpha radiation (E_p) was 694 10^{-4} μJ/m^3 with the range of 19-5584 10^{-4} μJ/m^3. The main parameter to be used in this analysis for describing the levels of radon in houses is the so-called equivalent concentration of radon (Ceq), whose values obtained from 185 measurements led to the following values: 12 Bq/m^3, 4.8 Bq/m^3 and 10 Bq/m^3 for the arithmetic mean, for the geometric mean and median, respectively, ranging from 0.5 to 100 Bq/m^3.

An independent series of measurements of radon concentrations in houses in the Białystok province was reported in [20]. Radon concentrations varied over the range of 1-1100 Bq/m^3, its arithmetic mean, geometric mean and median being 36.6 Bq/m^3, 18.4 Bq/m^3, and 17 Bq/m^3, respectively. To compare the results obtained with the use of both methods, it was necessary to adopt a coefficient of radioactive equilibrium in the air inside the dwelling houses investigated. In most cases, the value of this coefficient was found to be 0.4 [24]. When the above value and that of the mean equivalent radon concentration of 16 Bq/m^3 obtained by the Markov method are adopted for Białystok province [19], the radon concentration was estimated to be 40 Bq/m^3. This value is similar to the mean radon concentration determined in the same region with the use of environmental PICO-RAD detectors [20]. The mean radon levels in the whole northeastern part of Poland are lower than the corresponding mean radon value of 40 Bq/m^3 estimated for the world's population.

It is difficult to make comparisons between the above data and the current data reported by other researchers, since there is a tendency to avoid troublesome measurements of Rn-222 decay products in favour of much simpler techniques of direct radon measurements. The recent UN Report [12] gives the value of about 16 Bq/m^3 for the arithmetic mean of the equivalent radon concentration inside houses. The mean levels observed in northeastern Poland have never been found to exceed the above value of 16 Bq/m^3.

At present there is a possibility of appraising the results in light of legal regulations in force in Poland. The amendments adopted in July 1995 limiting exposure to radon and its products [25], introduced restrictions on the mean annual Rn-222 concentrations in premises occupied for permanent residence, abandoning the notion of ''equivalent radon concentration'' included in the Instruction issued by the President of the State Atomic Agency of 1988 [26]. An upper limit of 400 Bq/m^3 was adopted for radon concentrations in new buildings. From all the measurements carried out with both methods in the total of 324 houses, it was found that 99.4% of the results never exceeded the Polish norm (400 Bq/m^3), and higher levels were only noted in two uninhabited premises in cellars.

Apart from the measurements inside houses, other investigations were made at 15 points outside the buildings, giving a value of 4 Bq/m^3 for the mean radon concentration. This value is lower than that of 10 Bq/m^3 adopted as a mean radon level outside buildings in the world's continental regions [12].

Estimation of the Effective Dose Equivalent

From the results of the measurements of the dose rates from natural sources of ionizing radiation and from data in the literature concerning cosmic radiation, the average annual effective equivalent of the dose from natural environmental sources was estimated for a statistical inhabitant of ''Poland's Green Lungs'' region. The estimation was made for the population density of 1993 [1] using the following assumptions:

1. The average person in a moderate climate spends 20% of his/her time outdoors, and 80% indoors [3],
2. The mean dose rate from cosmic radiation, according to [27], was 36.4 nGyh^{-1}, allowing for ionization and neutron components and the correction for the variation of the dose with altitude,
3. On the basis of GUS (Main Statistical Office) data [28] and information from the building societies in the region, the following population distribution was assumed for houses constructed from the three typical materials: 15%, 25% and 60% in wooden, brick and prefabricated houses, respectively,
4. The absorbed dose in air was transformed into the effective dose equivalent with use factors 0.7 and 1.0 for gamma radiation and cosmic radiation, respectively [3],
5. The dose from radon was estimated assuming the time of staying indoors to be 7000 hr per year. The effective dose equivalent for radon products was estimated using the conversion coefficient of 10 nSvh^{-1}/Bqm^{-3} [24].

Table 1. Average annual effective dose to adults of northeastern Poland.

Lp.	Source of risk	q	Annual effective dose to adults mSv/year	Average value for Poland [29]
1	radon indoors	0.8	0.84	1.4
2	radon outdoors	0.2	0.01	0.06
3	gamma rays indoors	0.8	0.64	0.6
4	gamma rays outdoors	0.2	0.05	0.04
5	cosmic rays	0.2	0.07	0.07
	Total		1.61	2.17

q – coefficient of the time exposition

The annual effective dose equivalent per one statistical inhabitant of ''Poland's Green Lungs'' region was 1.61 mSv/year. The contributions from each component are shown in Table 1. The highest contribution in the effective dose equivalent (up to 53%) is due to radon in the air inside the houses. Table 1 gives a comparison of our data with those obtained by CLOR (Central Laboratory for Radiological Protection) for the whole of Poland [29]. When the same factors and mean values for Poland as those calculated by CLOR are adopted, the value obtained for the annual effective dose equivalent is 2.17 mSv/year, i.e. it is higher than that determined above. The main factor responsible for lower population exposition of the northeastern Poland is a lower radon level in indoor air.

The above analysis concerns natural ionization radiation sources in the environment. It follows that the inhabitants absorb a dose mainly when they are indoors. Thus, care should be taken in selecting proper building materials and construction designs to minimize this dose.

Estimation of Natural Radioactivity of Raw and Building Materials

In 1980 in Poland regulations concerning the content of naturally occurring radionuclides in raw and building materials were laid down with the purpose of limiting the increase in natural background gamma radiation inside dwelling houses resulting from the use of materials with higher levels of radioactivity [30]. Two preconditions were adopted to estimate the suitability of raw and prefabricated materials used in the construction of dwelling houses:

1. Coefficient f_1 which, as a condition of limiting the total body exposure to gamma radiation, should not exceed the value of 1, is given by

$$f_1 = 0.00027\ S_K + 0.0027\ S_R + 0.0043\ S_T < 1$$

where S_K, S_R and S_T are the concentrations of K-40, Ra-226 and Th-232, expressed in Bq/kg, in the samples.

2. Coefficient f_2, which is a condition of limiting the concentration of Ra-226 in the building materials due to the emanation of Rn-222 from walls, is given by

$$f_2 = S_R < 185\ \text{Bq/kg}$$

The unit that carries out investigations of the natural radioactivity of raw and building materials in northeastern Poland is the laboratory of the Department of Biophysics, Medical Academy in Białystok [31]. A total of 133 specimens, including 54 samples of raw materials and 79 prefabricated building products, were investigated. The results are given in Table 2.

The highest concentrations of Ra-226 and Th-232 were found in raw materials, where the mean values and the variation ranges (in Bq/kg) for Ra-226, Th-232 and K-40 were: 85 (26-150), 94 (16-911), and 599 (129-1032), respectively. Twice as low values were found for Ra-226 and Th-232 in bricks, i.e. 46 (26-65) and 51 (26-68). However, the values for K-40 were almost twice as great, i.e. 970 (411-1205). The best situation was observed in prefabricated products, where the mean concentration of radionuclides of radium, thorium and potassium was found to be the lowest, i.e. 25, 26 and 449 Bq/kg, respectively.

The assessment of the suitability of the materials investigated in light of the regulations in force was made in [15]. The results of that analysis are summarized in Table 3.

The most favourable situation was observed in prefabricated building materials, where the mean f_1 and f_2 coefficients had the lowest values, contributing only 33% to the permissible value of f_1 and 12% for f_2. In no case were the permissible values exceeded. Twice as high values were found in ceramic materials (bricks), although again they were not higher than permissible levels. Among the raw materials examined and used in the production of building materials, the mean f_1 and f_2 values contributed 66% and 42% of permissible values. In one sample of blast-furnace slag the f_1 coefficient was exceeded.

The total population exposure in Poland's northeastern region from environmental ionization radiation sources is presented in Table 4, where comparisons are made with the corresponding world data published in the UN Report [12].

Almost all the values found for Poland are lower than the world's mean values. In other words, the doses absorbed by the population in the northeastern region of Poland from natural sources of ionizing radiation are lower than the population-weighted world averages. This favourable situation of the radiological environment stands a chance of being preserved for the future. The region in question remains under the control of an ecodevelopment strategy that will undoubtedly restrict the negative effects of urbanization on the natural environment.

Table 2. Concentrations (in $Bqkg^{-1}$) of Ra-226, Th-232 and K-40 in building materials.

Kind of material	Number of samples	Ra-226		Th-232		K-40	
		Mean	Range	Mean	Range	Mean	Range
brick	37	46	26-65	51	26-68	970	411-1205
prefabricates	42	25	8-77	26	10-60	449	117-1179
raw material	54	85	26-150	94	16-91	559	129-1032

Table 3. Qualifying coefficients for building materials.

Kind of material	Number of samples	f_1		f_2	
		Mean	Range	Mean	Range
brick	37	0.6	0.29-0.74	45	26-65
prefabricates	42	0.3	0.12-0.75	23	8-77
raw material	54	0.66	0.24-1.00	78	5-154

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Table 4. The total population exposure in northeastern Poland from environmental ionization with comparisons to world data published in UN Report [12].

No.			Average value for northeast Poland	Population-weighted world average [12]
1	Absorbed dose rate in air from terrestrial gamma radiation ($nGyh^{-1}$)		40.2	57
2	Absorbed dose rate indoors-buildings made of: ($nGyh^{-1}$)	brick	102[a]	80
		prefabricates	89[a]	
		wood	88[a]	
3	Radon activity concentration indoors (Bqm^{-3})		30[b]	40
4	Radon activity concentration outdoors (Bqm^{-3})		4[c]	10
5	Mean activity concentration of natural radionuclides in soil ($Bqkg^{-1}$)	K-40	433[d]	500
		Ra-226	11[d]	40
		Th-232	15[d]	40
6	Mean activity concentration of natural radionuclides in building materials ($Bqkg^{-1}$)	K-40	693	500[e]
		Ra-226	34.8	50[e]
		Th-232	37.7	50[e]
7	Average annual effective dose to adults from radon (mSv)		0.85	1.2
8	Average annual effective dose to adults from cosmic rays and terrestrial radiation (excluding radon) (mSv)		0.76	1.07

[a] – measurements made with a use of ion chamber including gamma rays and cosmic rays indoors
[b] – mean value from introductory series (185 measurements) carried on Markov's method
[c] – mean value from measurements took in 15 points (open air)
[d] – mean value from measurements of 52 samples carried on 3 channel analyzer with NaI detector
[e] – mean activity concentration of natural radionuclides in typical building materials

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