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RADIUM AND RADON IN NATURAL UNDERGROUND WATER SUPPLY IN THE REGION OF ŁÓDŹ, POLAND

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Abstract. The region of Łódź, located in the central part of Poland, is very specific with respect to water supply coverage. In this densly populated and extensively industrialised region there is a lack of natural reservoirs of surface water in the nearest vicinity of Łódź. Therefore, over 50% of water for public and domestic use is provided from underground water intakes. It is well known from the literature that in underground water may occur radionuclides in the uranium and thorium decay series. Underground water intakes in Łódź used for public water supplies have been surveyed for the presence of radium-226 and radon-222 since the earlier studies of quality standards of the gross alpha and gross beta activity pointed to their possible excessive values. The surveillance was based on the guidelines set by the World Health Organisation (WHO). The measurements of radium-226 activity showed its different concentrations depending on the age of the water-bearing level. These concentrations fell within the range of $<10 \div 50$ Bqm^{-3}. Measurements of radon yielded similar results and its highest concentrations were found in the Lower cretaceous and Jurassic formations. Radon concentrations ranged from 200 to 11,400 Bqm^{-3}. A maximum annual dose taken into the body in the population drinking this water may account for 0.85 μSv which is lower than the limit recommended by WHO.

INTRODUCTION

Ionising radiation that originates from a number of naturally occurring sources in the environment constitutes a substantial constituent of human exposure, and contributes doses to the general population that are on average over two orders of magnitude higher than the existing limit for the exposure due to the application of radiation (28). Changes in the natural environment resulting from the process of economic, social and cultural development and its multifarious utilisation may frequently lead to an unnecessary exposure of the population to natural radiation.

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Water, particularly that from underground intakes, used directly for consumption or domestic purposes is one of the well-known sources of ionising radiation (22). The characteristic features of the Łódź region being the subject of our study are dense population, extensive industrialisation and lack of surface water settings. The urban water demand, totalling 260,000 m^3 per day, is provided from the Sulejów surface water reservoir on the Pilica river, and underground sources that supply 57% of water used. These data highlight the significance of the underground water intakes connected with a hydrological unit called the 'Łódź basin' for the city of Łódź. This water supply has been included into one of the 'main underground water reservoirs' and is under regular control and rigid protection.

Monitoring of water quality, involving the surveillance of physical and chemical water parameters at permanent measurement stations, is an essential element of the protection system. Monitoring is carried out in the so called 'observation networks' at the national, regional and local levels. For the monitoring purposes, the classification of water quality, including its radioactivity was verified in Poland in 1995 (26).

NATURAL RADIOACTIVITY IN WATER

The presence of radioactive elements, radium (^{226}Ra) and radon (^{222}Rn), in water was disclosed soon after the discovery of radioactivity owing to the studies of thermal sources carried out in England in 1904 (3). First comprehensive surveys of water radioactivity were initiated by the U.S. Environmental Protection Agency (U.S. EPA) in the 1950s (22). The surveys covered surface water, underground water and groundwater. An increasing interest in the studies of the occurrence of radionuclides in water has been evoked by:

(1) the growing application of ionizing radiation in various branches of the national economy, resulting in the elevated release of radionuclides to the environment;

(2) the guideline levels for radioactivity and admissible levels of the body intake for certain radionuclides laid down by the International Commission on Radiological Protection (ICRP) in the 1960s that helped to set their maximum admissible levels in different environmental media (10,11);

(3) the increased interest in problems of the human exposure to radon contributing mostly to doses received, and the search for exposure sources;

(4) the increasing difficulties in supplying water throughout the world, and a growing need to supplement the existing sources with underground water.

Two major sources of radioactive elements in water can be distinguished:

(1) infiltration of naturally occurring radionuclides in the uranium and thorium decay series in rocks and soils; and

(2) contamination of the natural environment resulting from man-made sources of radiation and radioactive wastes, mainly isotopes obtained artificially and nonexistent naturally, which are released to the environment.

The former source is responsible for contamination of underground water, and the latter contributes to the increased contamination of surface water. In order to limit contamination of surface water specific technical measures can be undertaken. In the event that natural elements are present in underground water their presence can be only reduced by surveying this water and eliminating water intakes known to be hazardous to human health.

Numerous studies have revealed that isotopes in the uranium series, mainly ^{226}Ra and ^{222}Rn, contribute mostly to natural radiation doses (27). The occurrence of these isotopes in underground water is primarily related to the geological make-up of an area where the water intakes are located. Generally, surface water is characterised by lower natural radioactivity. Surface water may be contaminated with these elements through a number of activities, including mining or using products with high natural radioactivity such as phosphate fertilisers (15,17,24). Average concentration of radium in surface water is about 10 Bqm^{-3}, and that of radon does not exceed several thousand Bqm^{-3} (1,7,9).

The studies carried out in various parts of the world indicate that these concentrations may be several dozen times higher in underground water than in surface water. For example, radon concentrations in some regions of the United States or Nordic countries reach the value of several $MBqm^{-3}$ (4,22).

The concentrations of this range create a significant problem in view of the protection of the population's health.

In Poland, the surveillance of underground water is haphazard and not systematic despite the fact that radiological quality factors have already been developed. The present situation in this regard can be attributed to certain problems concerning the methodology and equipment. For quite a long time it has been known that radon occurs in concentrations of 185 $kBqm^{-3}$ in water of some health resorts (Lądek and Świeradów). Drinking-water surveys for the presence of radon has been carried out only in the Mazowsze artesian basin (18) with Oligocenic water of high quality and good taste, very popular among inhabitants of Warsaw. In water from this basin radon concentrations vary, but they do not exceed 15 $kBqm^{-3}$.

POTENTIAL HEALTH EFFECTS RESULTING FROM CONSUMING WATER THAT CONTAINS NATURAL RADIONUCLIDES

Health effects which may be caused by consuming water that contains natural radionuclides can be considered in view of a dual pathway for exposure to these elements which present a potential hazard to the human body: (a) by ingestion from direct water consumption; and (b) by inhalation of the air containing water emitted radionuclides.

^{226}Ra isotope is the major source of the human body irradiation by radionuclides through the gastrointestinal tract. In areas with the natural radiation background, average annual absorption of ^{226}Ra accounts for 15 Bq from ingestion and of 0.01 Bq from inhalation (27).

The contribution of radioactivity absorbed usually with food is relatively low when consuming surface water but it may be significant for underground water.

Radium in the human body undergoes the same metabolism as calcium, and 70% of this element accumulates in bones, creating a permanent deposit, and becomes a source of the body irradiation (8). It is estimated that in areas of natural background radiation, an effective annual dose for a human being, resulting from intake of 15 Bq of radium, accounts for 7 μSv (27).

Exposure to radon contained in water is another problem. In this regard two issues should be considered: radon in water as a source of exposure of individuals by ingestion, and radon emanated from water as a source of exposure in the air.

Consumption of water containing dissolved radon leads to tissue irradiation with a specific dose which results mainly from alpha radiation emission from radon itself or its daughters. The majority of dissolved radon is released during water boiling process. A considerable amount of inhaled radon is quickly exhaled from the body via the lungs. Thus, doses originate mainly from radon decay products existing in water and produced during the presence of gas radon in the body.

It is thought that the greatest radiation dose, between 2 and 10 $nGyBq^{-1}$ is received by the stomach (27). Taking into consideration the radiation quality factor it may be assumed that the dose equivalent to the stomach will range from 40 to 200 $nSvBq^{-1}$, whereas 100 $nSvBq^{-1}$ is adopted as the average value resulting from numerous studies and models adopted. Based on the data presented, one may estimate that the consumption of 2 litres of water per day for 1 year contributes to the whole-body effective dose of 4 nSv per unit of radon concentration in water (Bqm^{-3}).

A model analysis of the effect of water-contained radon on the indoor air contamination may be based on the so called 'reference dwellings'. Assuming that radon concentration in water is 1000 Bqm^{-3} and that 50% of radon is emitted from water to the air, and providing that an average four-person family uses about 1.5 m^3 of water daily, it can be estimated that indoor radon concentration will increase by 0.1 Bqm^{-3} (27). These concentrations may considerably differ in time in individual households depending on the amount of water used. The aforesaid parameter of water use in an individual household adopted by the UN experts seems to be overestimated in Polish conditions. Nevertheless, it has been approved for further estimation of effects but it should be recognised as an upper limit.

Radon occurring in the air undergoes radioactive decay, develops a mixture of radon daughters which are heavy metals, and combined with air dust molecules produces radioactive aerosoles. This is most critical to the respiratory tract which becomes irradiated from radioactive deposit during the breathing process.

It is assumed that an effective annual dose contributed by the presence of radon in the air accounts for 17 μSv per the concentration unit (Bqm^{-3}) (27), which means that with the model use of water this dose contributed by radon inhalation is 2 nSv per water radon concentration unit (Bqm^{-3}).

RADIOLOGICAL STANDARDS FOR DRINKING-WATER QUALITY

First international standards for drinking-water, based on the ICRP recommendations were set by WHO in 1963. In 1984 this Organisation developed a system of guidelines for the quality of drinking-water with the aim of protecting health, and has recently revised these (1993). The guidelines are intended to provide the basis for national standards that will ensure the safety of drinking-water supplies by eliminating or reducing to a minimum concentrations of substances known to be hazardous to human health.

The recommended values are not intended to be used automatically as limits. They should be developed in the context of national or local environmental, social, economic and cultural conditions.

The guideline values provide a basis for assessing drinking-water quality, and their nature is as follows:

1. Setting values for concentrations of drinking-water constituents that does not result in any significant risk to the health of the consumer over a lifetime of consumption.

2. The quality of water defined by the guideline values is suitable for human consumption and for all usual domestic purposes, including personal hygiene.

3. When a guideline value is exceeded, this should be a signal to take remedial action and to consult the authority responsible for public health.

4. The guideline values are not intended to be used automatically as limits. It is essential that continuous efforts be made to maintain drinking-water quality at the highest possible level.

5. Short-term deviations above the guideline values do not necessarily mean that the water is unsuitable for consumption, but they should constitute an alarming signal to the authority responsible for public health, and encourage to take suitable action, taking also account of other sources of hazards, the toxicity of the substance, the likelihood and nature of any adverse health effects, and the effectiveness of remedial measures applied.

6. National standards for the quality of drinking-water should be based on the guidelines, but it is important that standards be developed in the context of geographical, social, economic, dietary and other conditions influencing potential hazards.

Based on the 1990 ICRP recommendations (13), the World Health Organisation has set the guidelines for radioactivity of drinking-water:

1. The recommended reference level of effective dose is 0.1 mSv from 1 year's consumption of drinking-water. This reference level of dose represents less than 5% of the average effective dose attributable annually to natural background radiation.

2. Below this reference level of dose, the drinking-water is acceptable for human consumption and any action to reduce the radioactivity is not necessary.

3. For practical purposes, the recommended guideline activity concentrations are:

– 100 Bqm^{-3} for the gross alpha activity, and

– 1000 Bqm^{-3} for the gross beta activity.

These recommendations apply to routine operational conditions of existing or new water supplies, and provide the basis for the control of quality standards for water.

Fig. 1 presents the procedures recommended by WHO to be applied in the event that the standards are exceeded (8).

WHO does not propose any recommendations for radon concentrations in water taking account of considerable difficulties resulting from the fact that radon is released from water during its treatment, assuming at the same time that in certain situations risk of exposure to radon by ingestion and inhalation may be comparable. However, WHO recommends to evaluate the effects of the presence of radon in water at the local and regional levels, taking account of other sources of radon existing in the human environment.

These effects have been assessed in some countries. For example, mortality from drinking radon-containing water in Sweden has been estimated by the Swedish Institute of Radiological Protection at 5–15 persons per year (2). The consumption of water with radon activity concentration of 1 $MBqm^{-3}$ may lead to absorption of quite high doses at the level of 7 mSv/year, this applies particularly to children who frequently drink tap water. Among adults this dose is estimated at 0.5 mSv/year (2).

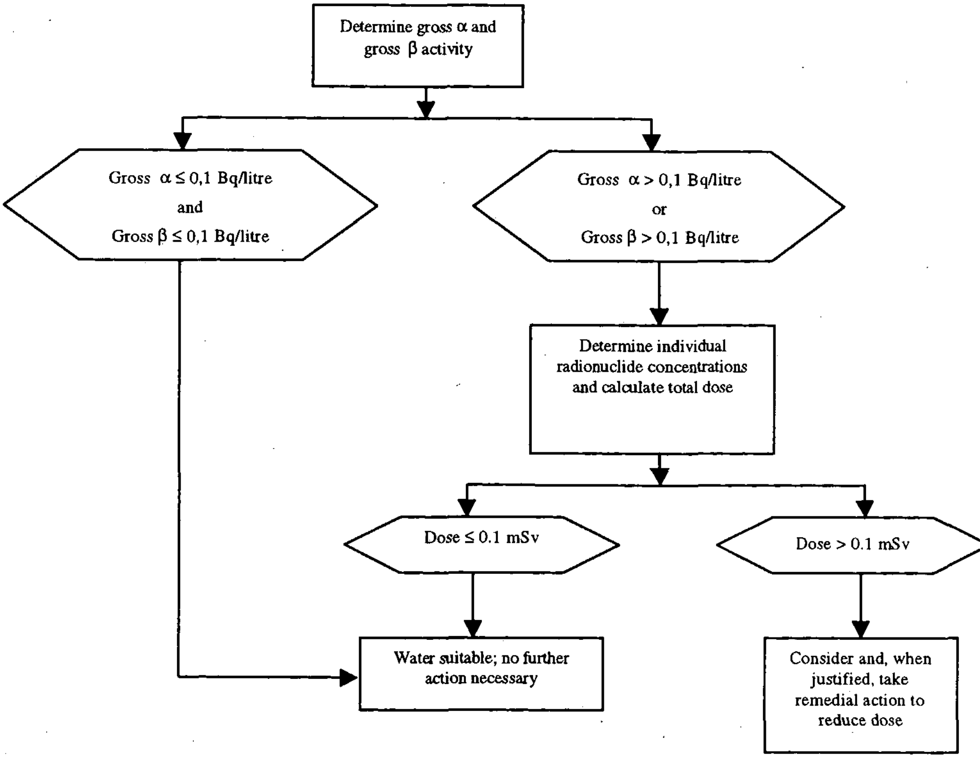

Fig. 1. The procedures recommended by WHO to protect against exceeding the guideline value of 0.1 mSv contributed by drinking-water.

Those studies provided the basis for setting in 1997 the reference level for radon activity concentrations in water. According to the legal regulations water containing more than 100 $kBqm^{-3}$ of radon requires treatment, and use of water containing more than 1 $MBqm^{-3}$ of radon is forbidden. These regulations apply both to public supplies and private wells (in Sweden there are about 200,000 private wells). The studies revealed that radon concentrations exceeded 100 $kBqm^{-3}$ in 50% of wells, and in 5% of them radon concentration exceeded 1 $MBqm^{-3}$. Consequently, the Swedish government has decided to provide the owners of wells with radon concentrations exceeding 1 $MBqm^{-3}$ with subsidies (5,000 SEC) for purchasing equipment necessary to reduce the exceeding concentrations.

Some reference levels based on the results from water surveys, carried out during the years 1978–1988, have been also adopted in the United States. The following recommendations related to the existing levels of concentration have been developed (5):

- concentrations below 37 $kBqm^{-3}$ – recognised as low levels that do not require repeated surveys;
- concentrations 37÷370 $kBqm^{-3}$ – possibly further surveys required;
- concentrations over 370 $kBqm^{-3}$ – further surveys required together with monitoring and water treatment.

In 1991, on the initiative of the Environmental Protection Agency, an attempt was made to set 'maximum level of water contamination' with radon at the value of 11 $kBqm^{-3}$. At the same time the basic requirements for water monitoring were laid down. However, in 1996 this attempt was relinquished, and the experts are now waiting for the estimation of costs involved in this kind of studies in the context of health risk reduction (16).

In Poland, the guideline levels for radioactivity in drinking-water has not as yet been developed (19), although hygiene standards for the production and distribution of mineral, spring and table water do exist (23). According to these standards the gross alpha radioactivity concentration was set at 100 Bqm^{-3}, and the gross beta activity concentration at 1,000 Bqm^{-3}, these values correspond with WHO guidelines. Quality factors for radioactivity of underground water adopted by the monitoring network are presented in Table 1.

Table 1. Classification of underground water quality for monitoring purposes

Indicator	Unit	Classification			
		I a the highest quality	I b high quality	II medium quality	III low quality
		I. Physical and physico-chemical indicators of water quality			
1. Gross alpha radioactivity	Bq/dm^3	0.05	0.1	0.2	0.2
2. Gross beta radioactivity	Bq/dm^3	0.5	1.0	2.0	2.0

The survey of the gross alpha and gross beta activity in 43 underground water wells in the city of Łódź revealed that the adopted value of gross alpha activity concentration – 100 Bqm^{-3} may be exceeded in some of them (29). In view of the WHO guidelines, the results obtained justify further studies aimed at identifying types of isotopes responsible for contamination.

Bearing in mind the aforesaid relationship between isotope concentrations and the volume of doses received by the population, the studies focus on identifying two essential isotopes in the uranium series: ^{226}Ra and ^{222}Rn.

MEASUREMENT METHODS

The methods for the collection and measurement of water samples met the world and national standards (6,20,21,25). Water samples were collected directly from underground intakes to Dreshl's glass bubblers, 250 ml in volume. The emanation method used involved the introduction of radon contained in water sample into scintillation chamber by bubbling the radon-free air through the bubblers. In the scintillation chamber, 6 dm^3 in capacity, two photomultipliers operating in the coincidence were installed. Electrode of negative potential placed inside the chamber was covered with ZnS (Ag) scintillator layer. This method increased the efficiency of the detection system by electrostatic collection of positively charged products of radon decay on the scintillator. The system calibration was based on standard radium solutions with activity of 1, 10 and 100 Bq produced and certified by the U.S. Isotope Products Laboratories. The lower detection limit for the applied measurement system was 10 Bqm^{-3}. The collected water samples were analysed twice. One measurement was performed directly after collecting water samples to determine radon activity. The second, performed 4 weeks later, was aimed to determine radon activity produced during radium decay in water sample. The measurement carried out in the state of equilibrium between these isotopes helped to define radium activity. The measurement uncertainty did not exceed 30% for the detection limit and was decreasing proportionally to increasing activity. The reliability of results was checked under the national system of comparative studies.

RESULTS

The survey covered underground water supplies used only for human consumption. In total, 43 underground water wells with four geological levels of water intakes: Quaternary, Upper createaceous, Lower createaceous and Jurassic, were surveyed. Only 10 intakes at the Quaternary level were used for local needs of inhabitants. Water from other intakes was supplied to central reservoirs where it was mixed with treated surface water. Table 2 presents the results from the analysis.

Table 2. An analysis of the measurement of radium and radon activity concentrations in water supplies at individual levels of geological formations

Meaurement	Evaluation parameter	Level of geological formations (no. of analyses)			
		Quaternary (20)	Upper createaceous (7)	Lower createaceous (8)	Jurassic (8)
Concentration of radium activity	Range (Bqm^{-3})	–	10÷13	10÷50	10÷23
	Mean (Bqm^{-3})	< 10	11	21	16
	Coefficient of variation (%)	–*	27	62	32
Concentration of radon activity	Range (Bqm^{-3})	1630÷7730	200÷4400	1070÷4690	3140÷11380
	Mean (Bqm^{-3})	3650	2930	2480	6700
	Coefficient of variation (%)	53	45	40	38

* concentrations in all samples studied were below the method sensitivity.

These results indicate the occurrence of elevated levels of radium concentrations in water originating from older geological formations (Lower createaceous and Jurassic) which is normal and correlates with earlier measurements of the gross alpha activity (29).

Similar observation was made in respect to radon activity concentrations, where the highest values were found in water originating from the Jurassic geological formations. These concentrations are close to those identified earlier in Oligocenic water in the Mazowsze basin (18).

CONCLUSIONS

Assuming that the highest detected radium activity concentrations account for 50 Bqm^{-3}, and providing that 2 litres of water is consumed per day, it may be calculated that the annual amount of this isotope taken into the body will equal 36 Bq, which means that the effective dose will be 17 μSv. This value exceeds over twice the average world level of a dose received by the population as a result of radium intake with food. However, bearing in mind that underground water originating from various sources is mixed before its supplying to the waterwork nets, it should be assumed that in reality water consumption will contribute to much lower effective doses. In addition, comparing the volume of this dose with that contributed by natural background and received *per capita* (2.6 mSv/year) it may be concluded that the aforesaid maximum dose from water consumption makes up only 0.6% of this dose.

Taking account of the effective-dose-coeficient of radon concentration in water, presented earlier, health consequences of consumption and use of water containing reference levels of radon activity concentrations can be estimated. For detected maximum concentration of 11,380 Bqm^{-3}, consumption of 2 litres of water per day will contribute to the effective dose of 45 μSv/year. Domestic use of water with such concentration in a typical dwelling of a four-person family may elevate radon concentration in the indoor air at home by 1 Bqm^{-3}, and the effective dose resulting from the body intake of radon by inhalation is about 23 μSv/year.

To sum up, the results from this analysis demonstrate that the maximum annual dose resulting from natural radioactivity in underground water used in the region of Łódź may account for 85 μSv/year, that is about 3% of the dose contributed by natural background, and lower than the guideline value of 0.1 mSv recommended by WHO.

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