Applied Radiation and Isotopes 93 (2014) 96–100

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

Natural radioactivity in drinking underground waters in Upper Silesia and solid wastes produced during treatment

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HIGHLIGHTS

- Analyses of water samples collected from underground wells usually show elevated natural radioactivity.
- The most crucial radionuclides are ^{226}Ra and ^{228}Ra.
- Attention should be focused on the fate of solid waste after water treatment.
- Radon in the air at water treatment plants should be monitored and to ensure compliance.

ARTICLE INFO

Available online 2 February 2014

Keywords:
Drinking waters
Annual effective dose
Solid waste materials
BCR procedure

ABSTRACT

Content of ^{226}Ra, ^{228}Ra and uranium isotopes in waters from subsurface aquifers was studied. The sampling points were chosen for having the elevated natural content of iron and manganese. Measurements of radium were made by LSC, while uranium was measured by alpha spectrometry. Waste sludge was measured by gamma spectrometry and three-stage BCR sequential extraction was performed. Radon activity concentration in the air at water treatment plants was determined and dose adsorbed by staff was calculated.

1. Introduction

In the most cases radionuclides present in water are of natural origin, mainly associated with uranium or thorium-bearing soils, mineral rocks. The occurrence of natural radioactivity in waters severely depends on the local geological structure of source, geochemistry of certain element and as well geochemical conditions in particular places (Ajayi and Achuka, 2009). The quality of drinking waters must be strictly controlled. For this reason drinking waters supplies, especially underground one, need to be monitored in terms of the among others natural radioactivity (Jobbagy et al., 2009).

The most studied radionuclides are ^{226}Ra, ^{228}Ra, ^{238}U, ^{234}U and ^{222}Rn because they may deliver the highest doses to people due to the consumption of waters. Radium enters groundwater by direct recoil across the liquid–solid interface between soil and water. After ingestion about 20% is initially distributed via blood to soft tissue, but finally it is retained in bone (de Oliviera et al., 2001). In case of uranium only 5% of ingested amount retains in human body, mainly in lungs and bone tissue. But rather the chemical toxicity of uranium as a heavy metal is emphasized, than risk arising from radioactive properties. It may cause kidney damage and problems with procreation.

Radon is water-soluble inert gas. Its high activity concentration is connected usually with granite rocks but also with karstic zones. Radon transported with water may lead to public exposure rather from transfer into the air in dwellings than from direct water consumption. The other places where increased radon exposure can be expected are water treatment plants (WTPs). Therefore special concern should be put on operators of WTPs.

Due to the occurrence of natural radioactivity in drinking water and the need for evaluation of potential exposure of the public, many international standards and guidelines have been issued. The World Health Organization (WHO) in “Guidelines for Drinking Water Quality” prior to qualitative water surveys recommends screening methods, namely measurement of gross alpha and beta activities. In the latest, 4th edition, these values were set 0.5 Bq/L and 1 Bq/L for alpha and beta activity respectively (WHO, 2011).

The European Commission published in 1998 Directive 98/83/EC on the quality of water intended for human consumption. In terms of radioactivity, only direct maximum acceptable value for tritium was set in that Directive. Content of the rest of radioisotopes is expressed as total indicative dose (TID) and should not exceed annual dose 0.1 mSv. TID excludes tritium, ^{40}K, ^{14}C, radon

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http://dx.doi.org/10.1016/j.apradiso.2014.01.017

and its decay products (European Commission (EC), 1998) and should be calculated accordingly to Council Directive 96/29 Euratom (European Commission (EC), 1996).

2. Materials and methods

Upper Silesia is located in the southern Poland (Fig. 1), covers about 7250 km^2. It is a large metropolitan area which consists of 37 towns, where 3.5 million of inhabitants live. Region is rich in mineral resources, mainly hard coal. Upper Silesian Coal Basin (USCB) is one from the largest in Europe. As a result of mining activity the region became highly industrialized and urbanized what in turn caused a large need for drinking waters. The primary surface water sources are Goczałkowice and Dzieckowice reservoirs. Research of those waters done in 2009 indicated that the level of natural and man-made radioactivity is very low and does not cause any radiological hazard for people (Chmielewska et al., 2011).

However, the vast majority of the region is supplied by drinking water from underground aquifers. Waters are drawn from depths range from 10 up to 300 m. Very often they contain considerable amount of iron and manganese and elevated level of natural radioisotopes. The most critical natural alpha and beta emitters in terms of internal contamination, ^{226}Ra, ^{228}Ra, ^{238}U, ^{234}U and ^{222}Rn are considered. Treatment is provided at the water treatment plants, using aeration, filtration and disinfection. Sporadically, coagulation by means of aluminium sulphate is included. This may cause the increase of radon activity concentration in air in closed rooms of WTPs.

Therefore in this work attention was focused on underground sources and the above mentioned radioisotopes. Water samples were collected within 2011–2012 years. Raw and treated waters were taken simultaneously.

The radiochemical procedure for ^{226}Ra and ^{228}Ra is exactly described by Chalupnik and Lebecka (1993). Radium isotopes were coprecipitated with barium carrier as Ba(Ra)SO_4, later on precipitate was dissolved in EDTA/NH_4OH in order to purify from other radionuclides (including ^{210}Pb). At pH 4.5 adjusted by glacial acetic acid, radium was reprecipitated with barium, whereas lead remained in solution as strong complex with EDTA. Ba(Ra)SO_4 was then mixed with gelling scintillator and measured 30 days later by means of LS spectrometer Quantulus.

Uranium was concentrated and isolated from waters by precipitation with $Fe(OH)_3$. Samples were spiked with a known amount of ^{232}U. Later the precipitate was centrifuged and dissolved in 9 M HCl. Soon after two columns were employed. The first one with anion exchanger (Dowex 1 × 8, 200–400 mesh) in order to remove thorium and other disturbing isotopes. The second column containing extraction chromatographic resin UTEVA was used for elimination of iron. Eventually, uranium was eluted with 0.02 M HCl and the alpha source was prepared by means of electroplating. The more detailed procedure for uranium separation is presented in Warwick (Warwick et al., 1999). Than alpha source was measured using high-resolution alpha spectrometry.

Track-etch detectors were used for measuring radon activity concentration in air of WTPs. They are CR-39 type detector foils in diffusion chamber. They were placed for 3 months in different WTP buildings in order to obtain mean radon activity in rooms, where water plant operators spent the most of their working time. Later on, CR39 foils were etched in 25% NaOH. And finally, the computer controlled optical evaluation system which counts the tracks on the detector foil corresponding to concentration of radon was used (Fisher et al., 1996).

As it was mentioned previously, in the most cases the sole applied method for water treatment turned out to be aeration in order to precipitate $Fe(OH)_3$ and MnO_2, accordingly with the following chemical reaction:

$$4Fe^{2+} + O_2 + 10H_2O \rightarrow 4Fe(OH)_3 + 8H^+$$

$$2Mn^{2+} + O_2 + 2H_2O \rightarrow 2MnO_2 + 4H^+$$

Sludge obtained during aeration and filtration processes adsorbs radionuclides on their surface and disposed in uncontrolled way could pose a serious problem for surrounding environment. In our study we collected sludge samples and after drying they were determined by gamma spectrometry. Finally in order to assess the potential negative impact of that sludge on natural environment the 3-stage sequential extraction according to BCR procedure (Ure et al., 1993) was performed. Scheme of that procedure is depicted in the figure below (Fig. 2).

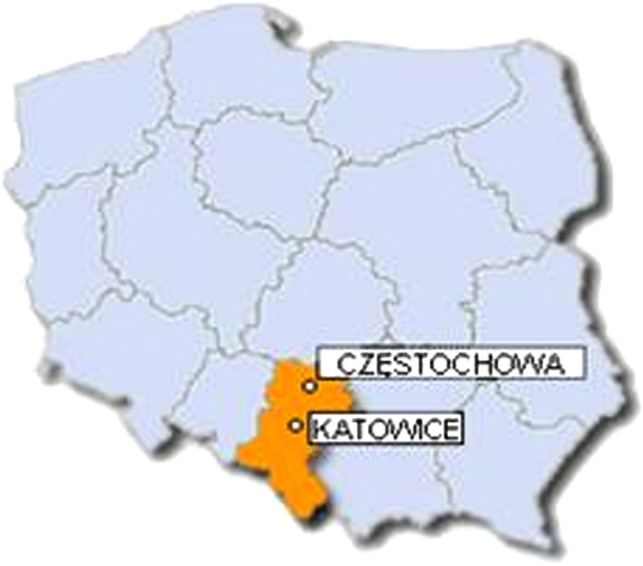

Fig. 1. Map of Poland, with marked Upper Silesia region, where drinking waters were monitored.

3. Results and discussion

Water samples were taken from 9 underground sources being within boundary of Upper Silesia region. Each time about 15 L of water was sampled, later on they were concentrated by evaporation and activity of certain isotopes were determined according to protocols mentioned before. Results are reported in Table 1.

The impact of radionuclides derived from drinking water can be assessed by determining the effective dose to each age group in population. The doses were calculated based on obtained activity concentration of radioisotopes for various water sources, assumed amount of water consumption for each age group (Table 2) as well dose coefficient recommended by Council Directive 96/29/Euratom (European Commission (EC), 1996).

Outcomes with the highest estimated annual dose are depicted below (Fig. 3). In many cases for raw waters the recommended dose 0.1 mSv/year was exceeded. Generally aeration used for removing iron and manganese caused decreasing of radioactivity. Nonetheless still are some treated waters, where despite of treatment process, annual dose 0.1 mSv is exceeded. The most crucial age groups are small babies, below 1 year and teenagers within 12–17 years.

In the next figure (Fig. 4) contribution of individual nuclides to total dose is shown. As an example we decided to present dose