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DETERMINATION OF STRONTIUM IN DRINKING WATER AND CONCEQUENCES OF RADIOACTIVE ELEMENTS PRESENT IN DRINKING WATER FOR HUMAN HEALTH

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Abstract: In this paper the analysis of strontium and uranium content in drinking water has been done, indirectly, according to the scale which originates from drinking water in water-supply system of the city of Belgrade.

Gamaspectrometric analysis showed the presence of free natural radionuclide in low activities. The activity of ⁹⁰Sr in scale which is 0.72±0.11 Bq/kg was determined by radiochemical. Because of the small quantities of fur in the house heater this activity can be considered as irrelevant, but the accumulation of scale can have intensified influence.

In this paper, the analysis of effects of the radioactive isotopes presence (first of all ²³⁸U and ²³⁵U) in drinking water on human health has been done.

Key words: drinking water, long lived radionuclides, uranium, strontium, scale, low-level ionizing radiation, kidney, chemical toxicity.

Introduction

The presence of radionuclide in drinking water can be result of various human activities. Radionuclide from nuclear fuel cycles and other (especially medical) usage can reach drinking water (Pavlović et al., 1995). According to

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the Manual of drinking water accuracy (Official Gazette of FRY, 1998), the maximum of allowed concentration of strontium in drinking water is not given.

Natural isotopes of strontium are not radioactive. Twelve other isotopes are, however, radioactive. Strontium-90 is artificial radionuclide which arises from the fission of U and Po, and in environment it is in larger concentration after the accident in Chernobyl. Half life of this isotope is 28.78 years.

Strontium-90 is highly radiotoxic for human beings because as chemical analogue to Ca it builds in human bones and teeth where it causes the damage of marrow and also leukemia (www.epa.gov).

The problem of analytical determination of isotope 90 Sr consists of separating from macro quantities of calcium and potential traces of barium. In literature different physical-chemical and physical methods for determination of this isotope are used. Physical-chemical methods are based on analytically chemical procedure with usage of collector (Шведов и Широков, 1962; Hallbach, 1959; Volchok, 1957; Linijecki, Czosnovska, 1960; Зацепина, 1966), then on ion-exchange chromatography (Pellerin et al., 1967; Porter, 1963; Davis, 1959; Tremillon, 1965) and extraction (François, 1965). Although these methods are used most often, isotope 90 Sr is possible to determin physically according to absorption of β- ray: $E(^{90}$ Sr) = 0.5 MeV; $E(^{90}$ Y) = 2.2 MeV (Шведо и Широков, 1962).

The aim of this paper was to determine strontium in drinking water indirectly, from the scale received by heating the water.

The usage of missile with different loading during NATO bombing of FR Yugoslavia in 1999 also warned of the possibility of presence of radioactive elements (mostly uranium and its isotopes) in drinking water (R a j k o v i ć, 2001) which could cause great consequences for human health, and this is also the subject of analysis in this paper.

Material and Methods

During the investigation, scale formed by precipitation on water heater surface over a 6-month period has been used. The scale originated from water present in water-supply system of the city of Belgrade - Pančevo-Krnjača. Content of total solids, which actually represents the scale, was determined by heating 1.0 dm³ of drinking water to boiling and evaporating it to obtain dry residue. A mass of dry residue was 0.2 g.

X-ray diffraction analysis has been performed by previously grinding of scale and the fraction of fine powder (6,3-2 μm) has been additionaly investigated. X-ray analysis has been performed on diffractometer Phillips PW 1009 with CuK α rays $\lambda=1.54178,$ at operating conditions of the tube U = 36 kV, I = 18 mA, with goniometer velocity Vg = 1°20/min with the conditions R/C = 8/2 (Martin, 1994).

Radiochemical method of separation 90 Sr is based on oxalate departing calcium from strontium, calcinating till oxide, and usage of tin as collector for 90 Y. The balance is established in 18 days, after which 90 Y departs on collector Al(OH)₃, which is then firing till oxide which is measured 90 Y after that (half-life period 64.2 hours) on α - β anticoincident counter with gas flow and low-phon (Countmaster-EG&G ORTEC). The size of planchete is 2.3 cm in diameter. The performance of counter is 24% and is determined by 90 Sr standard (Pantelić et al., 2004).

Gamma-spectrometric analysis of the scale was carried out so that the sample had been previously dried at 105°C (24 hours), in order to remove the moisture and reduce measurements to dry substance. Boiler fur was packed in small plastic vessels and hermetically closed to retain evolved radon. Measurement was done after 20 days, in order to balance developed radon with radium from which it originated.

Procedure for determination of 90Sr

The scale sample was dissolved in concentrated HCl with adding of distilled water. The solution was heated till 80°C and 8 wt.% oxalic acid in surplus was added. Then the solution concentrated NH₃ was added drop by drop till the pH 4.0 value. The sediment was left to rest during the night and then it was filtrated. The received sediment was heated at 800°C for two hours. Heated sediment was then soluted in HCl (1:1),30 mg aluminium and 2 g NH₄Cl was added to solution. Al(OH)₃ to which 90 Y was adsorbed, quantitatively sedimented with ammonia (pH = 7.1). The sediment is rejected after being washed with solution of NH₄Cl (2 wt.%) and distilled water. Aluminium in acid environment is added again to the filtrate. The solution was left to rest for 18 days till establishing radio active balance between 90 Sr and 90 Y.

Then aluminium hydroxide which was collector for the formed $^{90}Y(OH)_3$ was quantitavely sedimented again. This operation was repeated for the complete dividing and refinement of ^{90}Y from the traces of strontium and calcium.

Aluminium hydroxide sediment with filter paper was placed in measured porcelain bowl for heating, dried and burnt on flame, and then it was heated in electrical oven at 1200 °C at 10- minute intervals till constant mass.

The measurement of 90 Sr in scale was done through its radioactive descendent 90 Y on α - β anticoincident counter.

Results and Discussion

The results of the analysis of scale on atomic absorption spectrofotometer showed that in scale there were: calcium (as CaO) 48.85%, magnesium (as MgO)

4.24%, sodium (as Na_2O) 0.034%, potassium (as K_2O) 0.012%, iron (as Fe_2O_3) 4.02%, manganese (as MnO) 0.031%, silicon (as SiO_2) 0.10%, aluminium (as Al_2O_3) lower than 0.02%, uranium 1.1 ppm, while calcination loss was 42.67% (Rajković et al., 2004; Rajković, 2003; Rajković i Stojanović, 2001).

SEM photographs of heater scale samples are presented in Fig. 1.



Fig. 1. – SEM photograph of sample 1 of heater scale (x1.100)

The crystal structure of scale under electronic microscope indicates that scale is basically calcium carbonate that crystallizes hexagonally in the form of *calcite*, which was confirmed by making picture with diffractometer.

Diffractogram of the analysed scale sample is presented in the Fig. 2. In the obtained diffractogram the position of reflection was determined and <u>d</u> value of the measured area i.e. the integral intensities of certain reflections were calculated with the aid of the program DRX Win 1.4 designed by V.P.Martin (Martin, 1994).

Calcium carbonate (CaCO₃) in the form of *calcite* (crystalizing in hexagonal form) and *aragonite* (crystalizing in rombic form) gave numerous reflections in this diffractogram. Reflections of quartz are sharp and intensive. *Calcite* is detected on the basis of the strongest characteristic reflection at 3 Å and weaker ones at 3.8 Å, 2.47 Å, 2.26 Å, 2.07 Å and 1.89 Å. The presence of *aragonite* was established on the basis of weaker reflections at 1.74 Å, 1.84 Å, 1.98 Å, 2.11 Å, 2.34 Å, 2.7 Å, 3.29 Å and with the most intensive reflection at 3,42 Å.

On the basis of the presented diffractogram, it is not possible to establish the presence of other compounds, although an irregular and widened form of the reflection at 7 Å indicates presence of other substances, but at very low concentrations, which can not be determined by this method. This extended peak

indicates the presence of amorphous substances of organic and inorganic origin, and if inorganic, the most frequent of them is *limonite*.

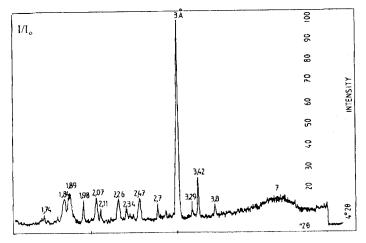


Fig. 2. - Characteristic X-ray diffractogram of the scale

The measurement time for each spectrum was about 55 hours, and all measurements were repeated after 20 days to obtain radioactive equilibrium of the evolved radon, and resulting spectrum is presented in Fig. 3.

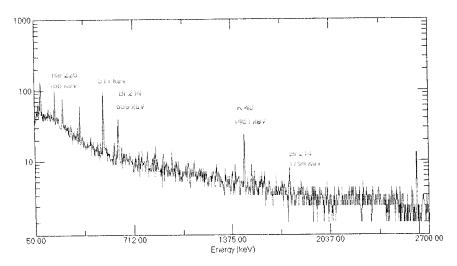


Fig. 3. - Gammaspectrometric spectrum of heater scale

The results of measurement of gamma-emitter activity concentration in the scale (in Bq/kg): 238 U (< 10); 235 U (<0.3); 226 Ra (4.6±1,3); 232 Th (1.8±0.5); 40 K (2.06±0.22); 134 Cs (< 0.2); 137 Cs (< 0.3). As natural uranium contains three isotopes: 234 U, 235 U i 238 U and in depleted uranium there is also 236 U, data show that uranium which exists in drinking water is of natural origin. Also, isotope relation 234 U/ 238 U in potable waters is 0.8-1.0, and values that deviate from this relation show the presence of depleted uranium. Using the method of fractional analysis (Rajković et al., 2005), it was found that uranium is mostly connected with mobile fraction – manganese and iron oxides (87.6 wt.%) ehich represents the fraction potentially available to human organism.

Calculated mass concentration of uranium in drinking water is $0.2 \,\mu\text{g/dm}^3$ (Rajković et al., 2004).

Gamaspectrometric analysis did not show the presence of any strontium isotope which is gama-emitt. The measured specific activity in the scale that comes from isotope ^{90}Sr is 0.72 ± 0.11 Bq/kg (Rajković et al., 2006; Pantelić et al., 2004). Calculated mass concentration of strontium in drinking water is $0.15~\mu g/dm^3$ (Rajković et al., 2004).

Strontium is found at a concentration far below that allowed by regulations for drinking water (2.0 mg/dm^3) (Official Gazette of FRY, 1998), but the presence of uranium should never be underestimated, nor considered to be negligible, knowing that it is highly toxic and radioactive element, which yields, through degradation, a sequence of radioactive daughters, in spite of comparatively high permitted concentration (0.05 mg/dm^3 i.e. 50 µg/dm^3) (Official Gazette of FRY, 1998).

The consequences of inter contamination with uranium

Uranium and its isotopes belong not only to radioactive elements, but also to highly toxic substances. Soluble uranium exerts the same toxic activity as soluble lead, but is 20 times more toxic than soluble tungsten (R a j k o v i ć, 2001). Considering this fact, the influence of uranium and its chemical effect as that of a toxic element is limited through the following concentrations (D o m i n g o, 1995; D o m i n g o et al., 1987; M a y n a r d et al., 1953; NIOSH, 1994):

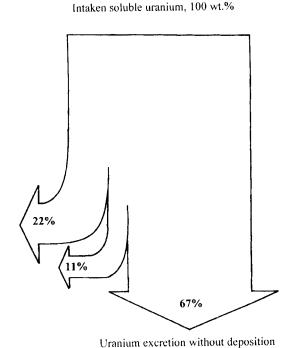
- in the air $-0.01 \text{ mg/dm}^3 (0.01 \text{ ppm})$;
- in the water -2.10^{-3} mg/dm³ (0.002 ppm or 2 μ g/dm³);
- the lethal dose amounts to 100 mg/kg body mass.

As a consequence of inter contamination of an organism by uranium, there arise certain risks and consequences for health that depend on 4 main factors: isotope content, chemical content, the size of a particle, and solubility. In the creation of pathogenic effect of uranium activity, chemical non-radiation toxicity and specific radioactivity of mainly gamma type (²³⁵U) and alpha-emission (²³⁸U)

have the same significance. Uranium kinetics in an organism is very specific. Blood is the main transport factor in distribution and redistribution of the most toxic uranyl (+6) and uranium (+4) ions. Plasma erythrocytes and proteins have the active role in transport. In 60 minutes 95% of reabsorbed uranium vanishes from blood and biological half life of uranium in the whole organism is 2-5000 days. Excretion is mainly by urine, and very small quantities from liver, through gall bladder and intestines, by feculence. The critical spot for soluble uranium compounds is kidney which is a predictable place of disposal. The amount from 0.05 to 12% is retained in kidneys with 6-1500 days half life and about 80% of total amount of uranium is exuded in 24 hours (Fig. 4).

Because of chemical toxicity of uranium in dissolved form, quantity of uranium of any isotope composition must not surpass in one day the limit of 2.5 mg uranium intaken from air (by inhalation) i.e. 150 mg uranium intaken through water or food (by ingestion) (Z a m o r a et al., 1998).

After kidneys uranium collects in skeleton, lungs, liver, spleen, pancreas, suprarenal gland and RES cells.



in any organ during first 24 h

Uranium deposited in bones

Uranium deposited in kidney, which is expelled with half life of 15 days

Fig. 4. - Flow sheet diagram of uranium distribution in human organism

Uranium is highly toxic for kidneys, while its radiation effect in only one intake is very weak. After only one intake and with well-timed therapy the changes are reversible, while at chronic exposure the alterations are generally permanent, thus leading gradually to the cessation of kidney functioning (Miller et al., 1998).

Taking into consideration the total mass of uranium that is taken in through the analysed drinking water of 0.3 mg (per I year), we come to a conclusion that the first 24 h 0.2 mg uranium is expelled from the organism, 0.03 mg is deposited in kindeys and 0.07 mg remain deposited in bones, from where it may be returned again into circulation and may be found in all parts of the body (Rajković and Stojanović, 2001).

Both pathogenic mechanisms: chemically-toxic and radiobiological, i.e. ionic that mutually interweave, insert and favour, are responsible for the arising of patho physiological mutations. As the result of uranium interaction with bio molecules in somatic cells and genetic material there arise numerous pathological effects that become visible very early or later effects visible after long lasting latent period (even up to 25 years) or even after few generations of genetic expression.

Uranium deposited in bones and other organs is subsequently returned into blood stream with at least two different half-lives, both longer than extraction half-life for kidney (ICRP, 1978; Alexander et al., 1988). Since uranium remains very long in bones, it exerts significant radiation effects. In kidneys and bone tissue U(IV) is primarily accumulated. In other tissues (liver, pancreas, spleen), 0.03-12% of intaken uranium is bound, mostly U(VI), before redistribution into kidney and bone system. Similarly to other heavy metals, e.g. mercury, uranium causes neurological problems, because it passes through a broken barrier (Rostker, 1998; Saladin, 1999).

People exposed to uranium have an increased risk of kidney functioning disturbance compared to those not exposed, and besides, before any changes in kidney functioning are noticed, 25% of kidney function may be lost, and even 75% may be lost before manifestation of actual clinical symptoms (SAIC, 1990).

The most serious and specific conditions arise as a consequence of mutagenic, teratogenic and cancerous effects. Genotoxic is the consequence of alpha radio active emission of uranium, which creates radiation cluster on chromatin in the nucleus of a cell. The increase of malign cancers arise 3 to 5 years after the first inter contamination with food and water.

Of numerous somatic disturbances the most important significance have the diseases of kidneys, stomach parahemat organs, suprarenal gland, disturbances of chematopoesae (blood), respiratory and neurological systems and immunological apparatus (Kovačević et al., 2004).

Conclusion

The analysis of scale originated from drinking water on the house water heater showed that scale consists mainly of calcium carbonate (*calcite*), which was confirmed by the electron microscope analysis.

The analysis of heavy metals content in scale, taken from water-supply system of the city of Belgrade, showed the presence of radioactive elements of uranium isotopes U-238 and U-235 specific activities in scale were below detection limit (less than I ppm). The presence of strontium was not confirmed by the examination on atomic absorption spectrophotometer (AAS).

Gamaspectrometric analysis indicates low activity concentrations of natural radionuclides. Activity concentration of ⁹⁰Sr obtained by radiochemical analysis is 0.72±0.11 Bq/kg.

Strontium is found at a concentration far below the one allowed by regulations for drinking water, but the presence of uranium should never be underestimated nor considered to be negligible, knowing that it is highly toxic and radioactive element, which yields, through degradation, a sequence of radioactive daughters, in spite of comparatively high permitted concentration.

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ODREĐIVANJE STRONCIJUMA U VODI ZA PIĆE I POSLEDICE PRISUSTVA RADIOAKTIVNIH ELEMENATA U VODI ZA PIĆE NA ZDRAVLJE LJUDI

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Rezime

U radu je izvršeno ispitivanje sadržaja stroncijuma i urana u vodi za piće, indirektnim putem, na osnovu kamenca izdvojenog iz vode za piće koja se nalazi u vodovodnoj mreži grada Beograda.

Gamaspektrometrijska ispitivanja ukazala su na prisustvo prirodnih radionuklida u niskim aktivnostima. Radiohemijskom analizom određena je aktivnost ⁹⁰Sr u kamencu koja iznosi 0,72±0,11 Bq/kg. Zbog male količine kamenca u kućnom bojleru ova se aktivnost može smatrati zanemarljivom, ali sa nagomilavanjem kamenca može doći i do pojačanog uticaja.

U radu je izvršena analiza posledica prisustva radioaktivnih izotopa (prevashodno ²³⁸U i ²³⁵U) u vodi za piće na zdravlje ljudi.

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