

NEW APPROACHES OF ESTIMATING URANIUM IN AIR

Yulia Iossifova¹, Rosen Rusev², Rosalina Chaturkova³

¹ M.D., Medical University of Varna, PO Box 177, Varna 9023, Bulgaria,
Telephone/Fax: (359) 52473304, e-mail: yuliayos@hotmail.com

² Institute of Hygiene and Epidemiology – Varna, Bulgaria, Tel: (359) 52332245,
GSM (359) 088 7602710

³ Technical University – Varna, Faculty of Marine Sciences and Ecology, Department
of Ecology and Environmental Protection, str. Studentska No 1, Varna 9010,
Bulgaria, Tel: (359) 52302085, e-mail: chaturkova@hotmail.com

Abstract

The methods of determining the background content of uranium in the atmospheric air in Bulgaria is mainly based on collecting samples with filters of perchlorine-vinyl fibres, burning of the filters in kilns and following estimation of uranium by standardised methods. This prolongs the measurement procedure significantly and decreases its exactness. We propose a new way for collecting samples – in water filter of volume of 2 litres distilled water by using the percussive -inertia method with vacuum absorbing system “Kerher”. The water filters provide higher speed of absorption and have greater dust capacity in comparison to dry filters. The sample collection lasts 7-8 hours with air volume capacity of 34 l/sec. The content of uranium in the water filter is estimated by the luminescent method. The background content of natural uranium in the atmospheric air in the town of Varna was determined – in one place for 3 months. The uranium concentrations vary from $2,5 \cdot 10^{-10} \text{ g/m}^3$ to $30 \cdot 10^{-10} \text{ g/m}^3$ and are significantly lower than the maximum allowed concentration. The research is a pilot one. Standardization of the method and enlarging the scope of the atmospheric air monitoring is a forthcoming process.

Introduction

The natural uranium is a mixture consisting of three isotopes: U^{234} (0,006%); U^{235} (0,7%) and U^{238} (99,3%), of which only uranium - 235 participates in the chain reaction of nuclear fission at its bombing with thermal neutrons (1). The process of disintegration of uranium is accompanied by emitting α - and β - particles, as well as γ - quanta. Nowadays uranium – 235 is the most important of all uranium isotopes. It is due to its capacity to divide under the impact of slow (thermal) neutrons. The fission of its nuclei by these neutrons is connected with the release of huge energy. That is why uranium –235 was first used for the production of nuclear bombs. At the end of the 50s years the weapons constructors came to the conclusion that uranium, 2,5 times denser than the steel, is eligible to pierce trough armored machinery. At the very moment of contact of the projectile with the armor, a very high temperature is reached immediately, resulting from the effect of the hit, as well as from the exothermal reaction between the uranium and the steel. In the space behind the armor the uranium, heated to very high temperatures, causes explosive reaction at the contact with air. It should also be added that the front armor of these tanks and

their gun-turret contain depleted uranium. According to the American specialists, its high density increases the defence capacity of armor. There are no uranium mines in Bulgaria, so that the only possible source of such could be a failure in the Atomic Electric Central and from 1999 military attacks in neighboring Yugoslavia, where bombs of depleted uranium were thrown in near proximity to the Bulgarian border. According to doctor Al-Yuesh, during the explosion part of the depleted uranium evaporates and is oxidized, which result is a very fine toxic ash. When this ash is inhaled in the lungs or enters the stomach, it emits α -particles, which are carcinogenic. Experiments with dogs (M.G. Tihaya et al) that inhaled uranium-235, showed development of lung malignancies, as a result of the radiation activity of α - rays. The quantity of uranium in environment at the use of armor-piercing missiles with shells of depleted uranium leads to radiation contamination of environment, after military attacks. The **aim** of the present work is the development of a method to measure the background content of natural uranium (U) in the atmospheric air, and thus to assess the environmental pollution. In Bulgaria the available data about pollution of the atmospheric air with uranium is scarce and is based on single observations, which are insufficient in order an assessment of the eventual risk to the Bulgarian population to be presented.

Methods

In the dry state uranium forms compounds of valences of 3+, 4+, 5+, or 6+. In aqueous media only U^{4+} and U^{6+} are stable. Some compounds, such as UCl_4 , decompose in aqueous media to the U^{6+} state. In acid solution and in the body, the oxygen-containing cation UO_2^{+2} , where uranium has a valence of 6+, is the predominant form. In general, hexavalent uranium compounds are the most soluble (2). The method used so far is not different from the proposed one, except that we use water filter and thus the process of uranium captivation is faster. The collection procedure was carried out, using the percussive-inertia method with vacuum absorbing system "Kerher". The content of uranium in the atmospheric air was determined for a period of seven hours (from 8 a.m. to 3 p.m.) during three consecutive months, year 2000. The capacity of the device for sample collection is 34 liters/second, which is equal to $857m^3$ air volume per sample. The sample of absorbed air is filtered through water filter of volume of two liters distilled water.

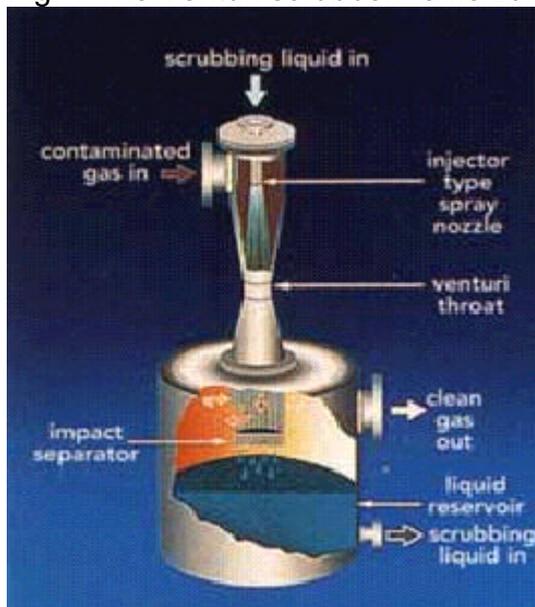
Table 1. A comparative characteristics of filters for aerosols

Type of the filter for aerosols	Velocity of the gas in the active zone, m/sec	Collection efficiency, % according the size of the particles (in microns)			Hydraulic resistance, kPa
		Up to 1	1-3	3-10	
Electric-type filters	0,3 – 1,5	75-95	90-99	98-100	0,1-0,3
Venturi Scrubber	50-150	90-97	95-100	98-100	5,0-20,0
Fibrous filters:					
- Low-speed	0,01- 0,1	92-99	96-100	100	0,5-5,0
- High-speed	1-10	50-85	85-97	95-100	1,5 –8,0
Netlike package	2,5 – 4,5	20-40	70-90	90-98	0,2-1,0

It is obvious from the table that the effectiveness of water filters to detain aerosols is comparable with the best filters of other types. The water filter provides high speed of the gas in the active zone, permanently great rate of air delivery. Unlike the dry filters, the water ones are of higher dust capacity. The advantage of water filters is that they are insignificantly influenced by the meteorological conditions, as well as

that there is no need to burn the filter in the kilns, which is time-profitable. In this research a Venturi scrubber with cyclonic droplet collector was used. A venturi scrubber utilizes the kinetic energy of an air stream to accomplish dust collection through the principle of impaction. The contaminated air stream is accelerated through a throat section reaching velocities between 45 and 120 meters per second. Water is introduced ahead of the throat and atomized by the high air velocity. Dust particles collide with and are captured in millions of small droplets. In order to avoid build-up of dust at the entrance of the venturi, a sharp wet/ dry zone is created. Water is injected tangentially around the top perimeter of the converging section to assure complete wetting of all surfaces. This design is known as the wet approach concept. In the long diverging section behind the throat, static pressure is regained as the velocity of the gas stream is reduced. The water – laden gas stream changes direction in a flooded elbow before entering the cyclonic separator tangentially where droplets are removed by centrifugal force. Water in the bottom of the elbow forms a cushion that prevents the high velocity water-laden gas stream from eroding the elbow itself. Clean, droplet-free air passes through the separator outlet and slurry is continuously drained from the bottom.

Fig. 1 The Venturi scrubber - a flexible wet dust collector



A venturi scrubber utilizes the kinetic energy of an air stream to accomplish dust collection through the principle of impaction. The contaminated air stream is accelerated through a throat section reaching velocities between 45 and 120 meters per second. Water is introduced ahead of the throat and atomized by the high air velocity. Dust particles collide with and are captured in millions of small droplets. In order to avoid build-up of dust at the entrance of the venturi, a sharp wet/ dry zone is created. Water is injected tangentially around the top perimeter of the converging section to assure complete wetting of all surfaces. This design is known as the wet approach concept. In the long diverging section behind the throat, static pressure is regained as the velocity of the gas stream is reduced. The water – laden gas stream changes direction in a flooded elbow before entering the cyclonic separator tangentially where droplets are removed by centrifugal force. Water in the bottom of the elbow forms a cushion that prevents the high velocity water-laden gas stream from eroding the elbow itself. Clean, droplet-free air passes through the separator outlet and slurry is continuously drained from the bottom. The content of uranium in

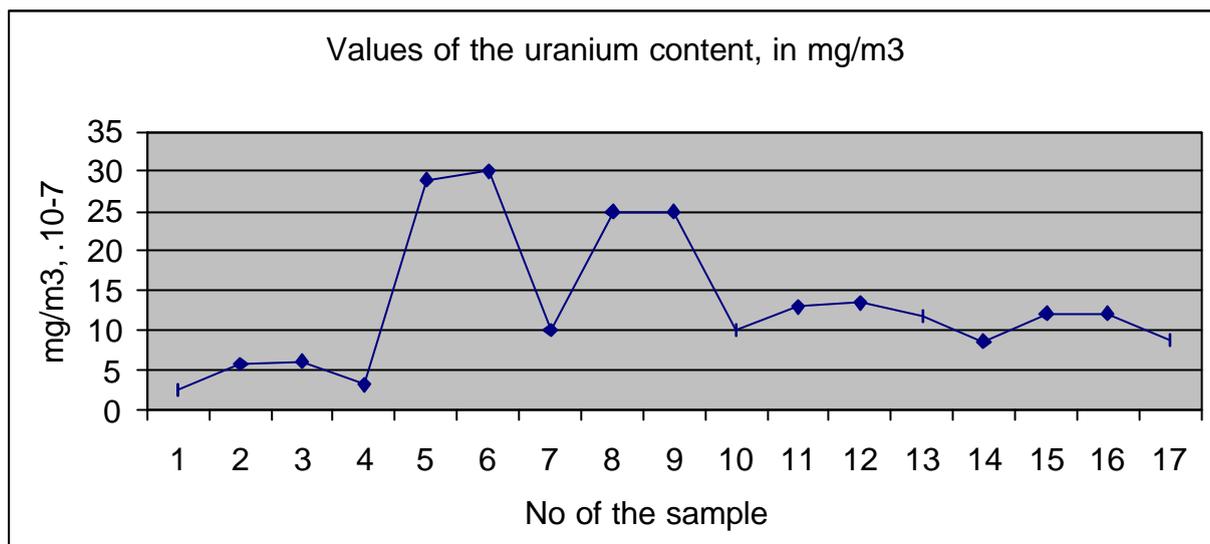
the water filter is determined by the “Luminescent method for determining the content of natural uranium in the drinking water”. The method is based on the nature of hexavalent uranium (U^{6+}) to luminescent with yellow-green light under the action of the ultraviolet light in melt with sodium fluoride. The light is proportional within certain boundaries to the uranium content. (Bulgarian Governmental Standard 12578-75, norm – 0,004 mg/m³ Uranium, soluble compounds). The used for this apparatus is: platinum and porcelain scale (dish), benzene burning instrument, Sokslet’s heater, quartz lamp, liquid extractor and platinum wire. The reagents used are as follows: double-distilled water, nitrogenous acid, ammonia nitrate, uranium oxinitrate hydrate, fluoride mixture, distilled ether and ammonia. The water in the sample of 1dm³ is being evaporated in a porcelain scale to dry in water bath. The dried remnant is processed with 10% nitrogenous acid in water bath to dry. The organic substances are burned out. The ash remnant is processed three times on water bath with 10% nitrogenous acid. Afterwards double distilled water is added, at which uranium passes to in a solution of uranium oxinitrate hydrate. The solution is put into the liquid extractor, after which 20cm³ double-distilled water and 150 cm³ distilled ether are added in the extractor container, and is constantly being extracted for three hours at fastly dropping of the ether. Afterwards the water extract containing the uranium is being evaporated to a small volume, and is transferred to a platinum dish, where it evaporates to dry. After the addition of a 100 mg fluoride mixture, everything is melted for 40 seconds at a flame. The melt is broken down and turned to a pearl. The pearl is irradiated with ultraviolet light and is compared with pearls-standards. Thus the content of uranium in the sample is established.

Results and Discussion

No of the sample	Its value in mg/m ³	Time in hours	Volume in m ³	Norms, mg/m ³
1	$2,5 \cdot 10^{-7}$	7	856,8	$4 \cdot 10^{-3}$
2	$5,8 \cdot 10^{-7}$	8	979,2	$4 \cdot 10^{-3}$
3	$6,2 \cdot 10^{-7}$	6.30	800,3	$4 \cdot 10^{-3}$
4	$3,2 \cdot 10^{-7}$	6.20	775,7	$4 \cdot 10^{-3}$
5	$29 \cdot 10^{-7}$	7.00	856,8	$4 \cdot 10^{-3}$
6	$30 \cdot 10^{-7}$	6.45	831,1	$4 \cdot 10^{-3}$
7	$10 \cdot 10^{-7}$	8.00	979,2	$4 \cdot 10^{-3}$
8	$25 \cdot 10^{-7}$	8.00	979,2	$4 \cdot 10^{-3}$
9	$25 \cdot 10^{-7}$	7.50	964,4	$4 \cdot 10^{-3}$
10	$10 \cdot 10^{-7}$	8.00	979,2	$4 \cdot 10^{-3}$
11	$13 \cdot 10^{-7}$	6.15	769,5	$4 \cdot 10^{-3}$
12	$13,5 \cdot 10^{-7}$	6.00	738,7	$4 \cdot 10^{-3}$
13	$11,9 \cdot 10^{-7}$	6.50	836,4	$4 \cdot 10^{-3}$
14	$8,7 \cdot 10^{-7}$	7.00	856,8	$4 \cdot 10^{-3}$
15	$12,2 \cdot 10^{-7}$	6.38	816,7	$4 \cdot 10^{-3}$
16	$12,2 \cdot 10^{-7}$	6.40	820,8	$4 \cdot 10^{-3}$
17	$8,8 \cdot 10^{-7}$	6.55	846,6	$4 \cdot 10^{-3}$

The results vary from $2,5 \cdot 10^{-10}$ to $30 \cdot 10^{-10}$ mg/m³, mean value of $13,35 \pm 8,34$ mg/m³.

The registered values are $3 \cdot 10^6$ times below the UPN (Upper Permitted Norm) for the population. The current report presents a pilot research of methods. We still have to establish the level of correlation with atmospheric conditions.



Conclusions

1. The results from the conducted control of the concentrations of radionuclides in air show values that are much below the UPN.
2. There is no risk of intake of radionuclids (U-234, U-235, and U-238) to the human body in the population as a whole.
3. The advantages of this method in comparison to the already practiced methods for sample taking and measurement of uranium in the atmospheric air in Bulgaria are as follows:
 - Mobility of the system;
 - The water filter provides a big and permanent rate of air delivery
 - Rational application of an already approved, standardized fluorescent method for establishing the uranium content in water, which provides for a short time the available uranium in the atmosphere to be registered.
 - This procedure is relatively quick, simple, and exhibits good reproducibility over the working range.
4. On the base of the results from the conducted measurements we propose a standardization and practical application of the present method and system.

References

1. Sahoo, S.K., Yonehara, H., Kurotaki, K., Fujimoto, K., and Nakamura, Y.: J. Radioanal. Nucl. Chem., **252**, 241-245, (2002).
2. Merck Index, 10th Edition, (1983), P. 9666.
3. I. Riekkinen, T. Jaakola, S. Pulli, S. Salminen, S. Ristonmaa, R. Rosenberg, R. Zilliacus. Analytical Methods for Wide Area Environmental Sampling (WAES) for Air Filters. STUK-YTO-TR 184, (2002).
4. W.C. Hinds. Aerosol Technology. Properties, Behaviour, and Measurement of Airborne Particles. 2. Ed. Wiley-Interscience, New York. 483 p., (1999)
5. J.G. Watson, J.C. Chow. Ambient Air Sampling. In: Willeke, K. and Baron, P.A. (Eds.) Aerosol Measurement: Principles, Techniques, and Applications. Van Nostrand Reinhold, New York. Pp. 622-639, (1993).
6. Valmari T, Tarvainen M, Lehtinen J, Rosenberg R, Honkamaa T, Ossintsev A, Lehtimaki M, Taipale A, Ylatalo S, Zilliacus R. Aerosol sampling methods for wide area environmental sampling (WAES), STUK-YTO-TR 183, (2001).