

CONTRIBUTION REGARDING THE RADIOACTIVE CONTAMINATION OF DRINKING WATER: HEALTH CONCERN, REGULATIONS, METHODS OF ASSESSMENT

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Abstract

The presence of radioactivity in drinking-water is a risk factor on human health, including cancer. This article presents the harmful effects of radioactivity on human health, the legislation and the available analytical methods of controlling radionuclides in environmental samples. Several case studies regarding the gross alpha and beta activities of surface, ground and drinking water, were described. According to the International Agency for Research on Cancer (IARC), radon, a radioactive gas that comes from disintegration of radium, is considered a carcinogenic agent of group I. There are two approaches of monitoring ²²²Rn in water, WHO and EURATOM. The methods for determination of radioactive content in water can be direct (gamma-spectrometry) or indirect (gamma-spectrometry, emanometry and liquid scintillation counting). Several published reports on radioactive pollution of water in different regions, showed exceeding values of gross alpha and beta activity, depending on geo-climatic factors. This study emphasizes the importance of monitoring water radioactivity and in particular radon, which can be a major risk for consumer health.

Key words: drinking-water, gross alpha activity, gross beta activity, natural and artificial radioactivity, radon.

INTRODUCTION

Environmental radiation is due to natural and artificial radionuclides. Radioactivity is present on earth, in different geological formations, in rocks, soil and water (Al-Khawlani et al., 2018). The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) has estimated that the global average annual human exposure to environmental radiation is about 3.0 mSv (UNSCEAR, 2008). Of this, 80% was due to natural sources of radiation, 19.6% to medical exposure and 0.4% was attributed to other man-made sources, e.g. nuclear power production and nuclear weapon testing (WHO, 2011; UNSCEAR, 2008).

Radiation occurs when energy is emitted by a source and then travels through a medium, until it is absorbed by matter (Sangiorgi et al., 2019). Natural radioactivity in water has been widely studied throughout the world in order to establish the radiological risk in humans

following consumption of contaminated water (Benedik, et al., 2012; Wallner et al., 2010; Labidi et al., 2010). In the last years, several radioactivity studies on water, soil and air samples have been conducted (Degerlier et al., 2010; Kam et al., 2010; Yarar et al., 2010; Kapand et al., 2012; Taskin et al., 2012), showing that levels of natural radioactivity in water can offer basic information on radiological hazards in drinking-water.

Radioactive materials ingested by humans may affect health as a result of the decay of radionuclides into the body. One of them is ²²⁶Ra, which is considered a highly toxic element for human (El-Gamal et. al., 2019). Environmental radionuclides could be absorbed and accumulated in certain organs or tissues causing potential risks for human health (Ogundare et al., 2015). There are studies suggesting that exposure to any dose of

radiations could induce cancer (Liang et al., 2015; Ogundare et al., 2015).

Considering these, there is a high requirement for quality and accurate radioactivity monitoring, in particular in water. Different analytical methods of measuring radioactivity from environmental samples have been described in the literature, such as gamma-spectroscopy (Bonotto et al., 2009), alpha-spectroscopy (Jobbagy et al., 2010) and liquid scintillation counting (ISO 11704 Water quality). As a result of the increased potential for radioactive contamination of water, a primary screening of gross alpha and gross beta measurements used as screening methods to detect changes of the radiological characteristics of drinking-water source, is required (WHO, 2011; Bunotto et al., 2008). Gross alpha and gross beta analyses are widely used as the first stage of radiological characterization of drinking-water (Jobbagy et al., 2014; Todorović et al., 2012; Cfarku et al., 2014; Jobbagy et al., 2010).

International standards and regulations impose permissible limits of the water radionuclides concentration and monitoring their levels using appropriate techniques (Rožmarić et al., 2012; Medley et al., 2015; Al-Hamarneh and Almasoud, 2018; Condomines et al., 2010; Diab and Abdallah et al., 2013; IAEA, 2014; Forte et al., 2018). However, the process of identifying procedures for evaluating the concentration of radionuclides of water samples is time-consuming and expensive. It has been shown that water physical-chemical properties are strongly related to the geological nature of the collecting site (La Verde et al., 2021). Therefore, the easiest practical approach is applying a screening method based on gross alpha and gross beta measurements, regardless of the identity of the specific radionuclides (QCVN 01-1:2018/BYT, 2018; WHO, 2017; Pintilie et al., 2016; Turhan et al., 2013).

THE IMPACT OF RADIOACTIVE CONTAMINATION OF WATER ON HUMAN HEALTH

Most radiations have their origin in the natural environment constituting the natural terrestrial background radiation. Thus, man has been exposed to the following natural ionizing radiation: (1) cosmic radiation – the amount (or

dose) of received cosmic radiation being influenced by altitude, atmospheric conditions and the magnetic field of Earth; (2) terrestrial radiation – due to radioactive substances (uranium, thorium and potassium) that exists in rocks, soil and water; (3) radon – radioactive gas element that exists in the environment (air, water) showing major contribution to the natural terrestrial background radiation (Burkhardt et al., 2016).

Radon (Rn) is a chemical element with atomic number 86, belonging to the group VIIIA. This noble gas is radioactive, tasteless, odorless and colorless. Therefore, it is not detectable by human senses alone. It is formed by the disintegration of the heavy elements from the Earth's crust. Once formed, it diffuses into the soil or water gases, being then emanated into the atmosphere. Radon migrates to the surface through the soil pores, fissures and erosions (Coretchi et al., 2020).

The access to a safe drinking-water is essential for the human health (Grande et al., 2015). The permissible radioactive levels of drinking-water are <0.5 Bq/l measured by gross alpha activity, and <1 Bq/l for gross beta activity (WHO, 2011). Additional investigation is required when levels exceed these limits (Cfarku et al., 2020). Regarding the ground water, the EU directives recognized about 1000 types of natural mineral ground water (European Commission, 2015). Considering the ground waters, the physical-chemical conditions and the geological environments strongly influence the level of radionuclides, higher contents affecting the human health by ingestion of drinking-water obtained from wells (Sarvajayakesavalu et al., 2018; Rozmaric et al., 2012; Altikulac et al., 2015).

Of all the dangerous radionuclides in water, radon (^{222}Rn) is of great concern, being produced by the decay of radium, the last one being the decay product of uranium (^{238}U). Radon is considered the main source of natural radioactivity with short-term products of disintegration of ^{238}U , including ^{214}Po , ^{214}Bi , ^{214}Pb and ^{218}Po (Richon et al., 2010; Binesh et al., 2012). Some radionuclides (^{228}Ra , ^{226}Ra , ^{210}Po) may accumulate in bones and teeth (La Verde et al., 2021).

According to the International Agency for Research on Cancer (IARC), radon is a

carcinogenic agent of group 1 (ICRP, 1988). Research indicated that inhaled radon may produce lung cancer, while ingested radon may produce gastric cancer (Binesh et al., 2012; Rafique et al., 2012). Considering smoking as the main risk factor of lung cancer, radon will be the first cause of cancer for non-smokers and the second one for smokers (Lorenzo-Gonzalez et al., 2019). The most amount of radon present in drinking-water is absorbed in the human body by inhalation and not by ingestion (La Verde et al., 2021). All radon isotopes are radioactive, so that the evaluation of adverse effects on human health due to radon exposure requires further consideration. The main health problems occur when the descendants of radon, which are attached to dust particles (called attached fractions) are inhaled, further deposited in

airways (tracheobronchial tree), thus repeatedly irradiating cells with alpha-particles as each atom suffers transformations through the disintegration chain. These alpha-particles provide a high dose of localized radiation (Keith et al., 2012).

LEGISLATIVE ASPECTS RELATED TO RADIOACTIVE SUBSTANCES IN DRINKING-WATER

²²²Rn in water – WHO and EURATOM perspective

Figure 1 presents the two approaches regarding the maximum allowed level of radon in water, according to The Guidelines for drinking-water quality (GDWQ) by WHO and EURATOM.

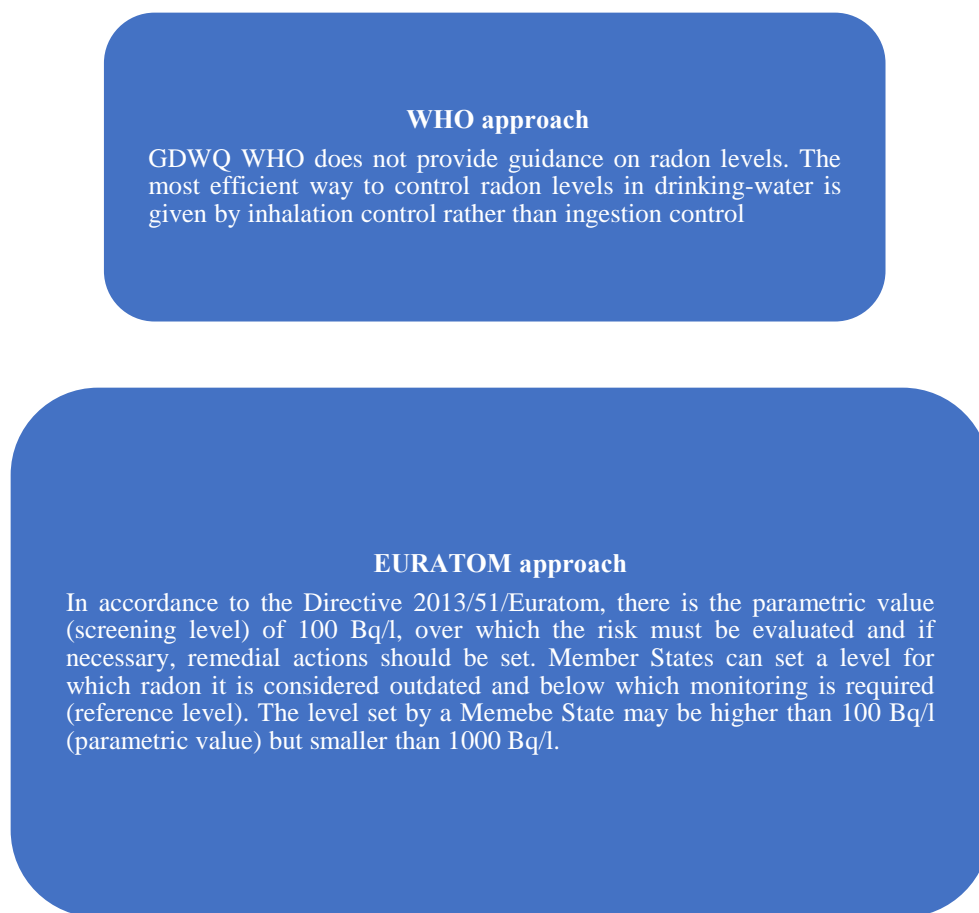


Figure 1. Comparative approach - WHO and EUROATOM – regarding the monitorization of radon (²²²Rn) in water

Gross alpha and gross beta activity in freshwater

The standard procedure describes the stages

required for the measurement of gross alpha and gross beta activity in freshwater. The collected stabilized water samples are evaporated and

subjected to drying, after which the sample residue is calcinated at 350°C for 1 h. The gross alpha and beta activity is measured from the water residue, results being obtained from a standard curve (SR-ISO 9696, 2018; SR-ISO 9697, 2019).

Health protection requirements in relation to radioactive substances in drinking-water

The national legal aspect (Law 301/2015) regulates the quality of drinking-water regarding the content of natural and artificial radioactive substances, by establishing the limit values, as well as frequencies and methods of monitoring radioactive substances in drinking-water, in order to protect the health of the population from the induced risk by the presence of radioactive substances.

According to the EU Council Directive 2013/51/EURATOM on the requirements for the public health protection against radioactive substances in water intended for human consumption, the maximum limit was set at 100 Bq/l (Council Directive, 2013). The WHO recommendation for the maximum level of ^{222}Rn in drinking-water is 100 Bq/l (WHO, 2011), while the U.S. Environmental Protection Agency (EPA) established the maximum level at 11.1 Bq/l (US-EPA, 1999).

CLASSICAL AND MODERN METHODS OF EVALUATION OF RADIOACTIVE CONTENT IN WATERS INTENDED FOR HUMAN CONSUMPTION

Because of the high cost of equipment used for measuring the radionuclides concentration, and

long analysis time, the easiest practical approach is a screening method based on radioactivity detection by gross alpha and beta activities (QCVN 01-1:2018/BYT; WHO, 2017; Pintilie et al., 2016; Turhan et al., 2013). Gamma-spectrometrical technique has been used to determine the specific activity of gamma-emitting radionuclides (anthropogenic and natural). The Liquid Scintillation Counting (LSC) method has been frequently applied to measure the activity of beta-emitting radionuclides (tritium, ^4C , ^{55}Fe , ^{63}Ni , $^{89,90}\text{Sr}$, ^{90}Y , ^{99}Tc , ^{241}Pu , ^{36}Cl , ^{41}Ca , ^{129}I , ^{210}Po , ^{210}Pb , isotopes of uranium, thorium, radium, and radon). The emanometry measurement technique has been used to estimate the activity of the gaseous radon (Caridi et al., 2021).

The methods used to evaluate the radioactive content of water samples are divided into the following categories (Bochicchio et al., 2019):

- direct measurement* without phase transfer: gamma-spectrometry;
- indirect measurement* involving the transfer of ^{222}Rn from the aqueous phase to gaseous phase, before performing the measurement: (a) gamma-spectrometry (radon adsorbed on charcoal); (b) emanometry, involving transfer of ^{222}Rn from the aqueous phase to gaseous phase; (c) LSC technique.

The description of the analytical methods, as well as advantages and disadvantages of each technique, is presented in Table 1.

Table 1. The main analytical techniques used to evaluate the radioactive content of water

Method type	Description	Advantages	Disadvantages	Ref.
Gamma spectrometry	The concentration of the ^{222}Rn is determined by measuring the characteristic gamma lines of ^{214}Bi or ^{214}Pb obtained by an HPGe (quantitative) or NaI (qualitative or semi-quantitative) detectors.	<ul style="list-style-type: none"> No sample treatment required; Data analysis is fully automatized; No specific training is required for the operators; Generally, the measurement uncertainty could be very low (< 5%) Corrections for the radon determination equation are required. 	<ul style="list-style-type: none"> HPGe detectors are highly expensive; High turnaround time, 4-13 h (few measurements/ week), compared other techniques The measurement results are influenced by indoor radon in the laboratory air. 	Bochicchio et al., 2019. Pujol et al., 2017

Emanometry	^{222}Rn is transferred from the liquid to the gaseous phase in a closed circuit by controlled sample degassing	<ul style="list-style-type: none"> • Different detectors coupled with the degassing circuit can be used, with low-to-moderate costs; • Measurement uncertainty can be very low (< 5%) if the method is properly managed; • Possibility to perform <i>in-situ</i> measurements; • Very low turnaround time, (<1h) => many measurements/day. • The equipment is portable • Rapid measurement 	<ul style="list-style-type: none"> • Degassing circuit required; • Sub-sampling is required: a certain quantity of water should be transferred from transport container to the degassing circuit; • The technique is sensitive to thoron (^{232}Th) 	Bochicchio et al., 2019 Caridi and Belmusto, 2018
Liquid scintillation counting (LSC)	The principle is based on the extraction of ^{222}Rn from water samples.	<ul style="list-style-type: none"> • The procedure is fully automatized; • Several vials can be analyzed at the same time => many measurements per day; • The lowest detection limit (0.05 Bq/l); • The vial to be measured can be prepared on-site: such procedure avoids the need of sub-sampling; • Indoor radon in laboratory air does not significantly influence measurement procedure and results. 	<ul style="list-style-type: none"> • Instruments for LSC are expensive; • The turnaround time is quite high (approximately the same as gamma-spectrometry), 3-8 h => no rapid results; • Calibration is cocktail specific, so each scintillation cocktail should be studied separately; • <i>In situ</i> measurements cannot be performed. 	Bochicchio et al., 2019

DETECTION OF RADIOACTIVE CONTAMINATION OF WATER USING GROSS ALPHA AND BETA ACTIVITY – CASE STUDIES

The most accepted protocol for radiological characterization of drinking-water consists in determination of the gross alpha and gross beta activities (Todorovic et al., 2012; Jobbagy et al.,

2012) in accordance to ISO standards for freshwater (ISO 9696:2018 and ISO 9697:2019).

The results of several reported investigations of radioactive contamination of surface and groundwater, for the period 2011-2020 synthesized from different international studies, are presented in Table 2.

Table 2. The results of published studies at national and international level regarding the radioactive contamination of surface and groundwater

Water samples	Origin country	Gross alpha activity (mean value) (mBq/l)	Gross beta activity (mean value) (mBq/l)	References
Groundwater	China (Haihe River Plain)	17-362 (112)	18-779 (171)	Yi P et al., 2018
Surface and groundwater	China	0.498-490 (29)	5-1260 (91)	Sang et al., 2020
Groundwater	North Vietnam	4.6-119 (38.7)	0.99-189 (88)	Duong et al., 2020
Groundwater	Iran (Guilan)	12-115 (52)	23-332 (110)	Abbasi et al., 2017
Groundwater	Iordan	180-9460 (1570)	360-7480 (1620)	Alomari et al., 2019
Surface and groundwater	Nigeria (Kaseno State)	24-665(142)	7-1330 (285)	Bello et al., 2020

Surface and groundwater	Nigeria (Kaseno State)	5.8-174	14.7-222.5	Fasae et al., 2015
Groundwater	Orwian / Nigeria	6.4-18.2	46-126	Ogundare, et al. 2015
Groundwater	Ado-Ekiti Metropolis	216-1299	64-582	Polytechnic et al., 2013
Surface and groundwater	Saudi Arabia	194	540	Amin et al., 2017
Groundwater	Hail/ Saudi Arabia	17-541 (215)	480-516 (260)	Shabana et al., 2014
Groundwater	Turkey (Nevşehir province)	13-182 (88)	81-779 (305)	Seref et al., 2019
Surface and groundwater	Serbia	1-13	41-173	Jankovic et al., 2012
Groundwater	Balaton/Hungary	35-1749 (189)	33-2015 (209)	Jobbagy et al., 2011
Surface and groundwater	Galati/Romania	<6.00-85.24 (22.18)	<25-434.85 (75.80)	Pintilie et al., 2016
Groundwater	Bucovina/Romania	0.40-45.40 (12.13)	1.51-47.45 (11.34)	Călin et al., 2016

The radioactivity evaluation of eight sources of thermal and drinking-water from North Vietnam (Duong et al., 2020) showed values of the determined gross alpha and beta activities between 38.7 mBq/l and 88.0 mBq/l. The minimum and maximum alpha and beta activity values were 4.6 mBq/l and 119.0 mBq/l, and 0.99 and 189 mBq/l, respectively. Lower values were reported in the study conducted in Iran (Guilan) (Abbasi et al., 2017) showing gross alpha and beta activity of 12 mBq/l and 115 mBq/l, respectively 23 mBq/l and 332 mBq/l. However, the values did not exceed the levels recommended by WHO: 500 mBq/l for gross alpha, and 1000 mBq/l for gross beta activity. A study conducted in China (Yi P et al., 2018) indicates values ranging from 17 to 362 mBq/l for gross alpha activity and, from 18 to 779 mBq/l for gross beta activity. These values were below the WHO allowed limits, in comparison with another study from China (Sang et al., 2020) in which increased gross beta activity (1260 mBq/l) exceeding the permissible limit was reported. The study conducted in Nigeria (Kaseno state) (Bello et al., 2020) reported

exceeding levels both for alpha activity (665 mBq/l) as well as for beta activity (1330 mBq/l). Similar increased values of gross alpha activities (1299 mBq/l) were found in Ado Ekiti Metropolis (Polytechnic et al., 2013). The highest values have been reported in Jordan (Alomari et al., 2019) for gross alpha activity (9460 mBq/l) and gross beta activity (7480 mBq/l), with an average value of 1620 mBq/l, and for Balaton/ Hungary investigation (Jobbagy et al., 2011) showing values of 1749 mBq/l for gross alpha activities, and of 2015 mBq/l for gross beta activities. The results of several studies of radioactivity of surface and ground waters conducted in Romania, in the regions of Galati (Pintilie et al. 2016) and Bucovina (Călin et al., 2016) indicated no exceeding levels. The highest value of gross alpha and beta activities were reported for samples collected from drilled wells in the study of Galati/ Romania.

Regarding the gross alpha and beta activities determined in drinking-water from different European regions, the results are presented in Table 3.

Table 3. The results of published studies regarding to radioactive contamination of drinking-water from different regions of Europe

Origin country	Alpha activity (mBq/l)	Beta activity (mBq/l)	References
Central Italy	18.18 – 128.18	41.57 – 258.59	Desideri et al., 2007
Spain	30-880	40-228	Palomo et al., 2007
Italy	8 - 349	25 - 273	Forte et al., 2007
Bulgaria	177	30 - 980	Kamenova-Totzeva et al., 2014
Portugal	15 – 330	18 – 457	Lopes et al., 2010
Greece	82	283	Karamanis et al., 2007
Albania	18 - 37	150-337	Cfarku et al., 2014

As noticed in Table 3, Spain reported values of gross alpha activity of 880 mBq/l, which exceeded the WHO recommended limit of 500 mBq/l. For the other European regions, values were within the permissible limits recommended by the WHO.

In all of these published studies, the variation of the values regarding the determined radioactive content of analyzed water samples is closely related to the different geological characteristics of the investigated area.

CONCLUSIONS

This article described the aspects regarding the impact of environmental radioactive contamination on human health, legislative aspects on monitoring the radioactive contamination in drinking-water, as well as specific methods for the evaluation of radioactive content of environmental samples.

The impact of natural/ anthropogenic radioactive environmental contamination on human health is related to several tissue injuries, including cancer.

The legislative aspects on monitorization of radioactive substances in drinking-water established their allowed limits, regulating the quality and safety of drinking-water.

Radionuclides analysis from water samples involves the use of different direct and indirect methods (gamma/ alpha-spectrometry, liquid scintillation counting). The easiest practical approach is a screening method based on radioactivity detection by gross alpha and beta activities.

Several case studies on gross alpha and beta activities have been presented, showing values which exceeded the permissible limits, which justifies the ongoing research in this field. Increase of gross alpha and beta activity above the reference level of 500 mBq/l and 1000 mBq/l, respectively, established by the WHO is due to the different geological characteristics, the properties of the soils and rocks specific to each region. The WHO recommendation for the maximum level of ^{222}Rn in drinking-water is 100 Bq/L, while the U.S. EPA established the maximum level at 11.1 Bq/l.

REFERENCES

- Al-Khawlany A. H., Khan A. R., Pathan J. M., 2018. Review on studies in natural background radiation. *Radiation Protection and Environment*, 41(4), 215.
- Abbasi A., Mirekhtari F., 2017. Gross alpha and beta exposure assessment due to intake of drinking water in Guilan, Iran. *J. Radioanal. Nucl. Chem.* 314:1075–1081.
- Altikulac A., Turhan S., H. Gumus, 2015. The natural and artificial radionuclides in drinking water samples and consequent population doses. *J. Rad. Res. Appl. Sci.*, 8, 578–582.
- Al-Hamarneh I.F., Almasoud F.I., 2018. A Comparative Study of Different Radiometric Methodologies for the Determination of ^{226}Ra in Water. *Nuclear Engineering and Technology* 50, 159–164.
- Alomari A.H., Saleh M.A., Hashim S., Alsayaheen A., Abdeldin I., Bani Khalaf R., 2019. Measurement of gross alpha and beta activity concentration in groundwater of Jordan: groundwater quality, annual effective dose and lifetime risk assessment. *J. Water Health*, 17:957–970.
- Amin R., 2017. Gross alpha and beta activities and trace elements levels in drinking water of Saudi Arabia Rafat Amin M *, *Adv. Appl. Sci. Res.*, 8: 62-69.
- Bello S., Nasiru R., Garba N.N., Adeyemo D.J., 2020. Annual effective dose associated with radon, gross alpha and gross beta radioactivity in drinking water from gold mining areas of Shanono and Bagwai, Kano state, Nigeria. *Microchem J.* 154:104551.
- Binesh A., Mowlavi A., Mohammadi S., 2012. Estimation of the effective dose from radon ingestion and inhalation in drinking water sources of Mashhad, Iran. *Int J. Radiat Res*, 10: 37-41.
- Bunotto D.M., Bueno T.O., 2008. The natural radioactivity in Guarani aquifer groundwater, Brazil. *Appl. Radiat. Isot.* 66:1507–1522.
- Bonotto D.M., Bueno T.O., Tessari B.W., Silva A., 2009. The natural radioactivity in water by gross alpha and beta measurements, *Radiat. Meas.* 44, 92–101.
- Bochicchio F., 2019. Management of radioactivity in drinking water including radon, Webinar on radon in drinking water organized jointly by IAEA and WHO.
- Burkhardt R., Dan T., Bogdan L., 2016. POPULATION HEALTH EDUCATION GUIDE, Ionizing Radiation Hygiene Laboratory, Cluj Regional Center for Public Health.
- Caridi F., Belmusto G., 2018. Radon radioactivity measurements in underground water: A comparison between different diagnostics technique.
- Caridi F., Pappaterra D., Belmusto G., and D'Agostino M., 2021. Radioactivity Measurements in Water: An Overview of the Actual Technologies, 17(6), 548-552.
- Calin M.R., Radulescu I., Ion A.C., Sirbu F., 2016. Radiochemical investigation on natural mineral waters from Bucovina region, Rom. *Journ. Phys.* 61, 1051–1066.

- Corețchi L., Bahnarel I., Gîncu M., Cojocari A., Hoffmann M., 2020. Control and assessment of the risk of population exposure to radon in the Republic of Moldova, *Medical Sciences*, vol.1, ISSUE 1.
- Cfarku F., Xhixha G., Bylyku E., Zdruli P., Mantovani F., Përpunja F., Callegari I., Guastaldi E., Xhixha M., Kaçeli, Thoma H., 2014. A preliminary study of gross alpha/beta activity concentrations in drinking waters from Albania. *J. Radioanal. Nucl. Chem.* 301:435–442.
- Cfarku F., Shyti M., Spahiu E., 2020. Gross Alpha/Beta Radioactivity In Drinking Water In The Main Cities Of Albania.
- Council Directive 2013/51/ EURATOM of 22 October 2013, Official Journal of the European Union pp. L.296/16.
- Condomines M., Rihs S., Lloret E., Seidel J. L., 2010. Determination of the four natural Ra isotopes in thermal waters by gamma-ray spectrometry. *Applied Radiation and Isotopes* 68, 384–391.
- Diab H. M., Abdellah W. M., 2013. Validation of ²²⁶Ra and ²²⁸Ra measurements in water samples using gamma spectrometric analysis. *Journal of Water Resource and Protection* 5, 53–57.
- Desideri D., Roselli C., Feduzi L., and Meli M. A., 2007. Radiological characterization of drinking waters in Central Italy. *Microchem. J.* 87, 13–19.
- Drinking water quality regulation 458/2002, republished. National legislation (OUG 86/2008).
- Degerlier M., and Karahan G., 2010. Natural radioactivity invarious surface waters in Adana, Turkey. *Desalination* 261 , 126–13.
- Duong H. V., Luong Le H., Duong T.N., Ngoc Minh V., Duong T.H., Hegedus M., Peka A., Kovacs T., 2020. Gross alpha/beta activity concentrations in spa and mineral waters in North Vietnam. *Journal of Radioanalytical and Nuclear Chemistry*, 326.2: 1511–1517.
- El-Gamal H., Sefelnas A., Salaheldin G., 2019. Determination of Natural Radionuclides for Water Resources on the West Bank of the Nile River, Assiut Governorate, Egypt, *Water*.
- EUROPEAN COMMISSION: LIST OF NATURAL MINERAL WATERS RECOGNISED BY MEMBER STATES, 2015.
- Fasae K. P., Ibikunle K. S.O., Akinkuade S.T., 2015. Gross alpha and beta activity concentrations in portable drinking water in Ado-Ekiti Metropolis and the committed effective dose, *Int. J. Adv. Res. Phys. Sci.*, 2, p.p. 1-6.
- Forte M., 2015. Validation of a method for measuring ²²⁶Ra in drinking waters by LSC. *Applied Radiation and Isotopes* 103, 143–150.
- Forte M., Rusconi R., Cazzaniga M. T., Sgorbati G., 2007. The measurement of radioactivity in Italian drinking waters, *Microchem. J.* 85: 98–102
- Grande S., Risica S., 2015. Radionuclides in drinking water: the recent legislative requirements of the European Union, *J. Radiol. Prot.* 35, 1–19.
- IAEA, 2014. A procedure for the rapid determination of Ra-226 and Ra-228 in drinking water by liquid scintillation counting, 7–12.
- International Commission on Radiological Protection (ICRP), 1988. Man-made Mineral Fibres and Radon. In *IARC Monographs on the Evaluation of Carcinogenic Risks to Humans*; IARC: Lyon, France; Volume 43, pp. 1–300.
- ISO 11704 Water quality: measurement of gross alpha and beta activity concentration in non-saline water—liquid scintillation counting method (under development).
- Jobbágy V., Merešová J., Wätjen U., 2014. Critical remarks on gross alpha/beta activity analysis in drinking waters: conclusions from a European interlaboratory comparison. *Appl. Radiat. Isot.* 87:429–4.
- Jobbágy V., Wätjen U., Meresova J., 2010. Current status of gross alpha/beta activity analysis in water samples: a short overview of methods. *J. Radioanal Nucl. Chem.* 286:393–399.
- Janković M. M., Todorovića D. J., Todorović N.A., Nikolov J., 2012. Natural radionuclides in drinking waters in Serbia, *Appl. Radiat. Isotopes* 70, 2703–2710.
- Jobbágy V., Merešová J., Wätjen U., 2012. Critical remarks on gross alpha/beta activity analysis in drinking waters: Conclusions from a European interlaboratory comparison, *Applied Radiation and Isotopes* 87, 429–434.
- Jobbágy V., Kávási N., Somlai J., Dombovári P., Gyöngyösi C., Kovács T., 2011. Gross alpha and beta activity concentrations in spring waters in Balaton Upland, Hungary, *Radiat. Meas.* 46, 195–163.
- Kam E., Bozkurt A., and Ilgar R., 2010. A study of back ground radioactivity level for Canakkale, Turkey. *Environ. Monit. Assess.* 168: 685–690.
- Kamenova-Totzeva R. M., Kotova, J. G., Tenev A. V., Totzev and Badulin V.M., 2014. Natural radioactivity content in Bulgarian drinking water and consequent dose estimation. *Radiation Protection Dosimetry*, pp. 1–6.
- Karamanis D., Stamoulis K. Ioannides K.G., 2007. Natural radionuclides and heavy metals in bottled water in Greece *Desalination*, 213, p.p. 90-97.
- Kapdan E., Taskin H., Kam E., Osmanlioglu A. E., Karahan G., and Bozkurt A., 2012. A study of environmental radioactivity measurements for Cankiri, Turkey . *Radiat. Prot. Dosim.*, 150 (3), 398–404.
- Keith S., Doyle J.R., Harper C., 2012. Toxicological Profile for Radon. Atlanta (GA): Agency for Toxic Substances and Disease Registry (US).
- Kobyay Y., Taşkın H., Yeşilkanat C.M., Çevik U., Karahan G., Çakır B., 2015. Radioactivity survey and risk assessment study for drinking water in the Artvin province, Turkey. *Water Air Soil Pollut* 226:49.
- Labidi S., Mahjoubi H., Essafi F., Bensalah R., 2010. Natural radioactivity levels in mineral therapeutic and spring waters in Tunisia. *Radiat. Phys. Chem.*, 79, 1196–1202.
- La Verde G., Artiola V., D'Avino V., La Commara M., Panico M., Polichetti S., and Pugliese M., 2021. Measurement of Natural Radionuclides in Drinking Water and Risk Assessment in a Volcanic Region of Italy, *Campania, Water* 13, 3271.

- Law 301/2015 on the stability of the requirements for the protection of the health of the population regarding radioactive substances in drinking water (Official Gazette no. 904 of December 7, 2015).
- Liang X., Song W., Li J., Jianxin Z., 2015. Research progress on drinking water radioactivity pollution and its health risk assessment 31, 30–32.
- Lopes I., Madruga M.J., Ferrador G.O., Sequeira M.M., Oliveira E.J., Gomes A.R., 2010. Monitoring of Gross Alpha, Gross Beta and Tritium Activities in Portuguese Drinking Waters. *Apartado* 21, 2686-953 Sacavem, Portugal: Estrada Nacional, 10.
- Lorenzo-Gonzalez M., Torres-Duran M., Barbosa-Lorenzo R., Provencio-Pulla M., Barros-Dios J. M., Ruano-Ravina, A., 2019. Radon exposure: a major cause of lung cancer. *Expert Review of Respiratory Medicine*, 13(9), 839-850.
- Medley P., Martin P., Bollhöfer A., Parry D., 2015. ²²⁶Ra and ²²⁸Ra measurement on a BaSO₄ co-precipitation source. *Applied Radiation and Isotopes* 95, 200–207.
- Ogundare F.O., Adekoya O.I., 2015. Gross alpha and beta radioactivity in surface soil and drinkable water around a steel processing facility. *J. Radiat. Res. Appl. Sci.*, 8:411-41.
- Palomo M., Pen˜alver A., Borrull F., Aguilar C., 2017. Measurement of radioactivity in bottled drinking water in Spain, *Appl. Radiat. Isot.* 65: 1165–1172.
- Pintilie V., Ene A., Georgescu L.P., Moraru L., Iticescu C., 2016. Measurements of gross alpha and beta activity in drinking water from Galati Region, Romania. *Romanian Reports in Physics*, Vol. 68, No. 3, P. 1208–1220.
- Polytechnic T.F., Nigeri E.S., 2013. Gross alpha and beta activity concentrations and committed effective dose due to intake of groundwater in Ado-Ekiti Metropolis; the Capital City of Ekiti State, Southwestern, Nigeria, *J. Nat. Sci. Res.*, 3, p.p. 61-67.
- Pujol L., Pérez-Zabaleta M. E., 2017. Comparison of three methods for measuring ²²²Rn in drinking water. *Journal of Radioanalytical and Nuclear Chemistry*, 314(2), 781-788.
- QCVN 01-1:2018/BYT. National technical regulation on domestic water quality, Ministry of Health (in Vietnamese) (2018).
- Rafique M., Manzoor N., Rahman S., Rahman S., Rajput M., 2012. Assessment of lung cancer risk due to indoor radon exposure in inhabitants of the state of Azad Kashmir; Pakistan. *Int J Radiat Res*, 10: 19-29.
- Richon P., Klinger Y., Tapponnier P., Li C-X, Van Der Woerd J., Perrier F., 2010. Measuring radon flux across active faults: Relevance of excavating and possibility of satellite discharges. *Radiation Measurements*, 45: 211-218.
- Rožmarić M., Rogić M., Benedik L., Štok M., 2012. Natural radionuclides in bottled drinking waters produced in Croatia and their contribution to radiation dose. *Science of the Total Environment* 437, 53–60.
- Sangiorgi M., Hernández Ceballos M.A.H., Iurlaro G., Cinelli G., and Marc de Cort, 2019. 30 years of European Commission Radioactivity Environmental Monitoring data bank (REMdb) – an open door to boost environmental radioactivity research.
- Sang C., An W., Sørensen P. B., Han M., Hong Y., Yang M., 2021. Gross alpha and beta measurements in drinkable water from seven major geographical regions of China and the associated cancer risks. *Ecotoxicology and Environmental Safety*, 208, 111728.
- Shabana E.I., Kinsara A.A., 2014. Radioactivity in groundwater of high Background radiation area. *J. Environ. Radioact.* 137, 181–189.
- Sarvajayakesavalu S., Lakshminarayanan D., George J., Magesh S.B., Anilkumar K.M., Brammanandhan G.M., Chandrasekara A., Ravikumar M., 2018. Geographic Information System mapping of gross alpha/beta activity concentrations in ground water samples from Karnataka, India: a preliminary study. *Groundw Sustain Dev.* 6:164–168.
- SR. ISO 9696: 2018- “Calitatea apei. Măsurarea activității alfa grosse la apa nesalină”.
- SR. ISO 9697: 2019- - “Calitatea apei. Măsurarea activității beta grosse la apa nesalină”.
- Şeref T., 2019. The natural radioactivity in drinking water by gross alpha and beta measurements and radiological quality assessment. *Radiochim Acta*.
- Taskin H., Kam E., and Bozkurt A., 2012. Determination of gross alpha and beta activity concentrations in drinking waters in Bursa region of north-western Turkey. *Desalination Water Treat.* 45, 21–25.
- Todorović N., Nikolov J., Tenjović B., Bikit I., Vesković M., 2012. Establishment of a method for measurement of gross alpha/beta activities in water from Vojvodina region, *Radiat. Meas.* 47, 1053–1059.
- Turhan S., NAEM, Zriba H., Taşkın Z., Yılmaz S., Bayülken A., Hançerlioğulları A., Kurnaz, 2019. Radiochemical analysis of bottled drinking waters consumed in Turkey and a risk assessment study. *Microchem J.* 149:104047.
- Turhan S., Özçtak E., Taşkın H., Varinlioğlu A., 2013. Determination of natural radioactivity by gross alpha and beta measurements in ground water samples. *Water Res.* 47:3103–3108.
- United Nations Scientific Committee on the Effects of Atomic Radiation UNSCEAR, 2008. Report to the General Assembly, with scientific annexes, Vol. 1. Sources and Effects of Ionising Radiation.
- US EPA (United States environmental protection agency) 1999, USEPA (United States Environmental Protection Agency). Radon in drinking water health risk reduction and cost analysis Federal Register, Washington, pp. 9559-9599 Vol. 64.
- Wallner G., Jabbar T., 2010. Natural radionuclides in Austrian bottled mineral waters. *J. Radioanal. Nucl. Chem.*, 286, 329–334.
- WHO, 2011. Guidelines for drinking-water quality. Chapter 9 Radiological Aspects, 4th Edition. World Health Organization Library Cataloguing-in-Publication Data NLM classification: WA 675, Geneva.
- WHO, 2017. Guidelines for drinking-water quality 4ed Ch. 9, 203–218, WHO publications, Geneva.
- Yarar Y., Kam E., and Bozkurt A., 2010. A study of background radioactivity level for Terkirdag Turkey (1), 40-44.

Yi P., Gong M., Zhang W., Hou X.L., Aldahan A., Yang J., Chen P., 2018. Evaluation of gross- α and gross- β activities in groundwater of the Haihe River Plain, China. *J. Radioanal. Nucl. Chem.* 317:193–201.