IOSUD – UNIVERSITATEA „DUNĂREA DE JOS” DIN GALAȚI

Doctoral School of Mechanical and Industrial Engineering

UNIVERSITAS GALATIENSIS

Ph.D. THESIS

ABSTRACT

CONTRIBUTIONS REGARDING TO POPULATION'S EXPOSURE TO IONISING RADIATIONS

Ph.D. Student,

Violeta NICOLOV (Pintilie)

Scientific coordinator,

Prof. Univ. Dr. Ing. PUIU LUCIAN GEORGESCU

Seria I4: Inginerie Industrială Nr. 58

GALAŢI

2019

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Violeta NICOLOV (Pintilie)

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Industrial Engineering: I4 Nr. 58

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Second prize - Scientific Conference of the Doctoral School of "Dunărea de Jos" University of Galati, 4-th edition

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PARTICIPATION IN NATIONAL AND INTERNATIONAL PROJECTS:

1. Programul Operational Comun "Bazinul Mării Negre" 2014-2020, Contract de grant nr. 105067/14.09.2018, Proiect "***Creating a system of innovative transboundary monitoring of the transformation of the Black Sea river ecosystems under the impact of hydropower development and climate change***" - ***HydroEcoNex*** – cod BSB165, (expert);

2. Programul de cercetare, dezvoltare şi inovare pentru sistemele fluvii, delte, mări – Danubius, Codul proiectului: 4/07.05.2018, Titlul proiectului: ***Strategie şi acţiuni pentru pregătirea participării naţionale la Proiectul DANUBIUS–RI***, DANS;

3. Programul: ***Rețeaua de cooperare interdisciplinară în Bazinul Mării Negre pentru monitorizarea comună durabilă a migrației compușilor toxici în mediu, evaluarea îmbunătățită a stării ecologice și a impactului substanțelor dăunătoare asupra sănătății umane și prevenirea expunerii populației – MONITOX,*** cod BSB27 **(**voluntar);

4. Proiect nr. 80/2017: ***Investigation of advanced functional materials using atomic and nuclear analytical technique and imaging microscopy,*** Tema 03-4-1128-2017/2019 din planul tematic IUCN Dubna, Ordin IUCN nr. 220/10.04.2017 Dubna – Director proiect: Prof. Dr. Habil. Antoaneta Ene; (member);

5. Proiect nr. 81/2017: ***Applied research on air and soil pollution with toxic elements using nuclear and related analytical techniques***, Tema 03-4-1128-2017/2019 din planul tematic IUCN Dubna, Ordin IUCN nr. 220/10.04.2017 Dubna – director Proiect: rof. Dr. habil. Antoaneta Ene; (member);

6. Grant IUCN Dubna nr. 24/2016, Ord. IUCN nr. 94 din 15.02.2016, (Tema 03-4-1104-2011/2016), ***Development of infrastructure of spectroscopy and microscopy laboratories used for the characterization of environmental and crystalline materials***, voluntar, Director Grant IUCN Dubna: Prof. Dr. Habil. Antoaneta Ene; (member);

7. Proiect IUCN Dubna Nr. 104/2016, Ord. IUCN nr. 96 din 15.02.2016, (Tema 03-4-1104/2011/2016), ***Investigation of crystalline materials (diamonds and boron nitrides) using atomic and nuclear analytical techniques and imaging microscopy***, voluntar, Director Proiect Prof. Dr. Habil. Antoaneta Ene; (member);

INTRODUCTION

The doctoral thesis entitled "***Contributions regarding to population's exposure to ionising radiations***" had the objective of quantifying the main sources of exposure of the population to ionizing radiation through the ingestion of drinking water and food, on the one hand and by inhalation air in public and living spaces, on the other hand.

For this purpose, radioactivity parameters of drinking water, mineral water and food were determined, from the regions of Galați, Brăila and Vrancea counties, between 2015-2018. These data were used to evaluate *the annual effective dose* due to the ingestion of natural radionuclides through the consumption of water and food, after that the detriment brought to health was evaluated, due to ionizing radiation. Also, the concentration of radon and thoron in the air of the public and living spaces was determined and on the basis of it the annual effective dose due to the inhalation of the air from these spaces was evaluated.

Chapter 1 of the thesis presents state of art from of scientific literature data regarding:
- the main ways of exposing the population to ionizing radiation,
- dosimetric sizes and units,
- methods used to determine the radionuclides investigated in the present study,
- characterization of radionuclides determined in this study,
- techniques for determining the radioactive content of water and food,
- legislative regulations regarding the radioactive content in drinking water and food.

These aspects were tracked in view of the radioactivity indicators determined in the present study, from water and food, respectively: global alpha and beta activity, as well as natural radionuclides ^{210}Po, ^{210}Pb, ^{nat}U, ^{nat}Th, ^{226}Ra, ^{40}K.

Chapter 2 presents the methods used in this study to determine the parameters of the radioactivity to be assessed from proposed drinking water and food. It also specifies the data on way the samples were collected and the sampling area.

Chapter 3 presents experimental data on the following areas of research:
- exposure assessment population to ionizing radiation due to ingestion of natural radionuclides through consumption of drinking water,
- evaluation of population exposure to ionizing radiation due to ingestion of radionuclides through the consumption of food: bread, meat, pasteurized milk, milk powder for children from the age category menu for 0-1 year, early childhood 2-7 years (Galați, Brăila și Vrancea counties),
- evaluation of population exposure to ionizing radiation by ingesting radionuclides ^{210}Pb, ^{210}Po, ^{226}Ra through consumption of mineral water,
- evaluation of population exposure to ionizing radiation due to radon in the air of residential and public spaces.

This chapter begins with the presentation of methods of assessing the *Annual effective dose* due to ingestion of radionuclides through consumption of drinking water and food. This because, in the literature, several methods are used for evaluating the *Annual effective dose* received by the population through ingestion of radionuclides. Of these, some may overestimate, others may underestimate actual dose assessment. The chapter ends with the presentation of the findings resulting from the primary experimental study about the topic addressed in this thesis.

Chapter 4 presents the most important interpretations and conclusions drawn from experimental data carried out in the framework of this thesis, as well as assessments the detriment due to ionising radiations brought public health through the consumption of water and the food. They are also exposed to opportunities of expansion of the theme addressed in future research directions.

The thesis has been carried out with the guidance of the scientific commission;:

- Prof. Ph.D. Eng. PUIU-LUCIAN GEORGESCU – Scientific coordinator,
- Prof. Ph.D. Eng. LUMINIȚA MORARU,
- Prof. Ph.D. Eng. Habil. ANTOANETA ENE,
- Prof. Ph.D. Habil. CĂTĂLINA ITICESCU.

Research results have been presented at three national conferences and nine international conferences. The research results were also disseminated in five scientific papers published in ISI journals and four scientific papers published in BDI journals. The scientific published papers have received a total of eight citations (in journals: *Journal of Radioanalytical and Nuclear Chemistry, International Journal of Ambient Energy, IOSR Journal JAP of Applied Physics, Romanian Journal of Physics, Journal of Bioscience, Biochemistry and Bioinformatics, Networking of Mutagens in Environmental Toxicology, Environmental Geochemistry and Health*, Water and Conference Proceedings: *RAD Conference Proceedings*) [Abbasi A., 2017], [Oghenevovwero E. Esi, 2018], [Abbdy A. G. E., 2017], [Timofti M., 2019], [Ion I., 2019], [Gupta M., 2019], [Călin M. R., 2019], [Aydogdu M. H., 2019] (http://www.rad-proceedings.org/index.php?id=2, 2017).

The motivation of the thesis:

The national and international studies have shown correlations between certain diseases and radioactivity, generally. A very important component is the natural radioactivity, therefore the idea of this thesis was to concentrate the researches and their interpretation through the complex data regarding the natural radioactivity. The topic of the thesis "Contributions regarding to population's exposure to ionising radiations" is the evaluation of the exposure of population to ionizing radiation due to ingestion and inhalation of the natural radionuclides. This study quantifies the *annual effective dose* for the population of the counties of Vrancea, Brăila and Galați, through consumption of drinking water and the foodstuffs, during the period from 2015 to 2018. These administrative-territorial units were chosen for the study, because they present varied geological characteristics of the soils of provenance of the water sources used in the process of making water, in order to be distributed as drinking water to the population. The elements evaluated from the point of view of the radioactive content are: drinking water, mineral water, as well as basic foodstuffs: bread, milk and meat. For example, in milk the radionuclides are quickly transferred from the environment (one chain: feed-animal-milk), thus being a very good indicator of environmental radioactivity. Bread - as an element of study of quantification of the *annual effective dose*, was chosen due to the fact that in Romania the bread it is commonly used in alimentation. The radionuclides ^{210}Pb, ^{210}Po, ^{nat}U, ^{226}Ra, ^{40}K, ^{nat}Th were chosen due to their committeed effective doses per unit intake - notion explained in *Chap 3.*

The data obtained in this thesis can be used in subsequent observation of changes of water content and radioactive food.

It was also evaluated the *annual effective dose* due to inhalation of radon from the air of public buildings and dwellings ifrom Galați county, since it represents 43% of the contribution of ionizing radiation exposure to the population.

Estimation of population exposure to ionising radiation has been made in accordance with the recommendations of the International Commission of protection: Radiological (ICPR) and the United Nations Scientific Committee on the effect of ionizing radiation (UNSCEAR).

Key words: gross alpha/beta activity, specific activity/concentration of ^{210}Pb, ^{210}Po, ^{nat}U, ^{226}Ra, ^{40}K, ^{nat}Th, Galați, Brăila, Vrancea, drinking and mineral water, foodstuffs, annual effective dose, detriment

1. STATE OF ART

1.1. The pathways to exposure to ionizing radiation of the population

United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) estimates, at the global level, that the contribution of the natural sources to the annual effective dose is 2.4 mSv year^{-1}; from this, 1.2 mSv is due to Inhalation (mainly radon), 0.5 mSv to terrestrial gamma radiations, 0.4 mSv to cosmic rays and 0.3 mSv to ingestion of food and water [UNSCEAR, 2000].

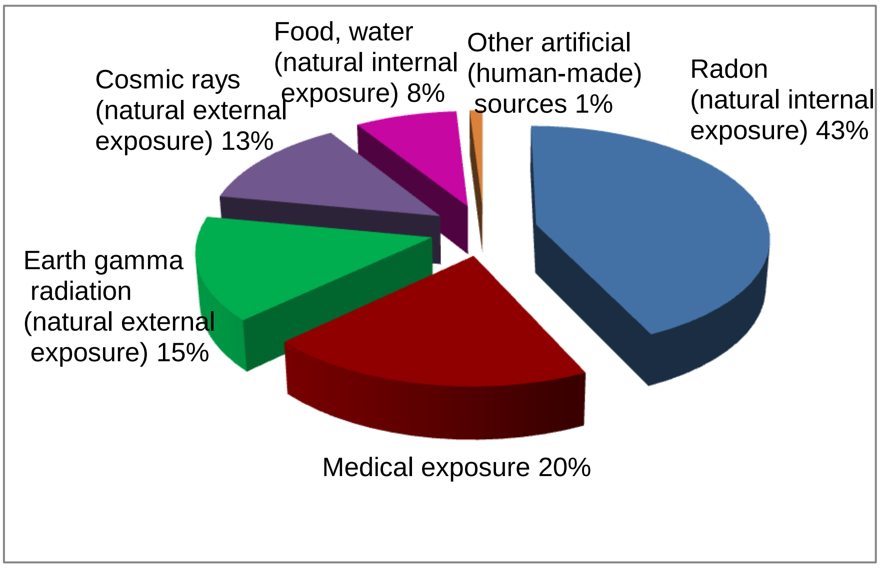


Figure 1.1 *– Radiation exposure pathways for the population and their contribution to the effective dose*

The annual effective dose is a measure of all the biological effects of ionizing radiation on the human body [Chiosilă I., 2014]. The annual effective dose for people in the population is the sum of the effective dose of one year due to external and internal exposure during the same year. *The effective dose of radiation received by a person is, in simple terms, the sum of the equivalent doses received by all tissues or organs, weighted for "tissue weighting factors". These reflect different sensitivities to radiation of different organs and tissues in the human body. The unit for the effective dose is J kg^{-1}, and its special name is sievert (Sv)* [WHO, 2008], [*ICRP, Publication 103*]..

The most important pathways of the exposure to ionizing radiation of the population are exemplified in *Figure 1.1.*

The natural radionuclides from drinking water come from three naturally radioactive series: thorium series, actinium series and uranium series. In the case of natural radionuclides of uranium and thorium series, the effective dose conversion factor by ingestion for adults grows in the following order:

$$^{238}U<^{235}U<^{234}U<^{224}Ra<^{226}Ra<^{210}Pb<^{228}Ra<^{210}Po$$

Uranium, radium, and thorium occur in three natural decay series, headed by uranium-238 thorium-232, and uranium-235, respectively.

The uranium series is exeplified in *Figure 1.2.*

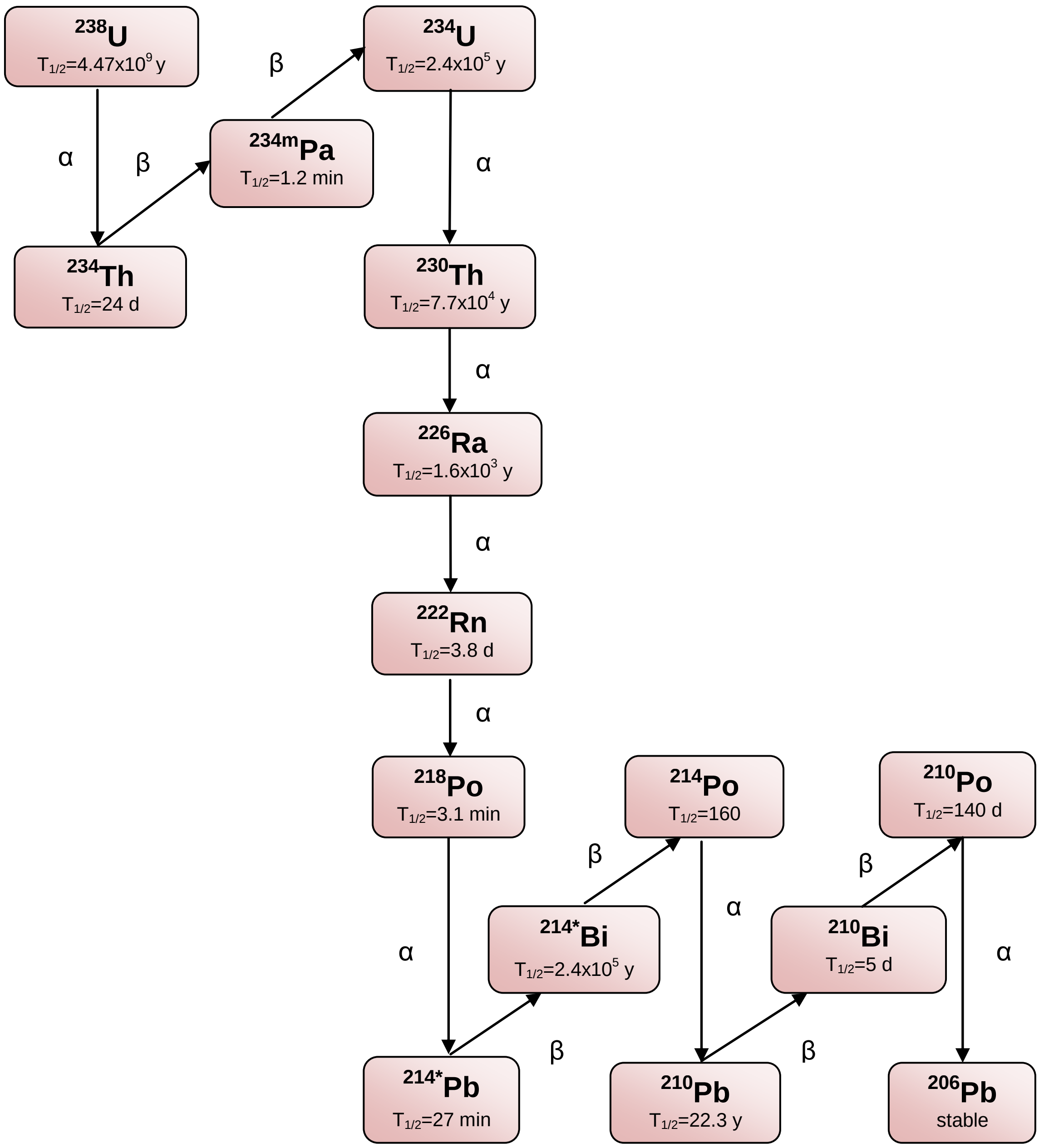


Figure 1.2 *- Natural Decay Series: Uranium-238 (4n+2)*

Water is an indispensable component in our lives and the managing of the water resources is a national and international problem. Water bears the radioactive, microbiological and chemical fingerprints of the crossed environment, being a very important transporter of these elements. A major influence on the composition and mobility of components from water has the regional geology [Spanos T., 2015], [Calin M. R., 2016]. The quality of the water destined to human consumption must be strictly controlled in a certain region as regards the physico-chemical, biological and radioactive parameters [Spanos T., 2015], [Calin M. R., 2016], [Calmet D., 2013], [Murkren Al-J. H., 2015].

Consumption of food, water and air inhalation represent the most important processes which contribute to the internal exposure of the human body to the ionizing radiation.

The alimentary tract is an important pathway of the ingestion of radionuclides. The contribution to the dose due to ingestion of natural radionuclides depends to the rate of intake, to the concentrations of radionuclides, and to the conversion factor from activity to dose. In order to evaluate the effective dose due to ingestion of radionuclides it is necessary to determine their concentration in food [Solecki J., 2011], [Štrok M., 2011], [Pintilie V., 2017], [Pintilie V., 2018a], [Pintilie V., 2018b], [Ene A., 2017], and water [Rožmarič M., 2012], [Gorur F. K., 2014], [Ogundare F. O., 2015], [Pintilie V., 2016a], [Pintilie V., 2016c], [Pintilie V., 2017a], [Abassi A., 2017].

1.2. Description of the radionuclides determined in the present study

Polonium-210:

One of the most radiotoxic alpha radionuclides characterized by high committed effective dose per unit intake via ingestion is polonium-210 [UNSCEAR, 2000]. Due to high relative energy (5,3 MeV), the polonium-210 tends to concentrate in the soft tissue, muscle, liver and causes sclerotic changes of blood vessels [Kristan U., 2015], [Štrok M., 2011]. As a result of polonium-210 determinations on the human brain, the victim of Alzheimer's disease, [Momčilović, 2006] it was observed that the polonium-210 it is accumulated in the hippocampus and tonsils. It has been previously observed [Söremark, 1966] that after intravenous injection of mice with polonium-210, this did not accumulate in the brain, suggesting that Alzheimer's disease was not related to the direct ingestion of radionuclide polonium-210, but rather to radon disintegration products. (radon-222) [UNSCEAR, 2000].

***Table 1.3** – Description of the polonium-210*

Description	Numerical value
Atomic mass, A	209,98 g/mol
Atomic number, Z	84
Half life, $T_{1/2}$	138,38 days
Belong to the radioactive series	the radioactive series ^{238}U
Daughter-nuclide	^{206}Pb
Decay mode	100% alpha
Maxime Energy	5304,33 keV (100%)
Specific activity	44,98 10^2 Ci/g

Lead-210:

One of the most radiotoxic beta radionuclides is lead-210. This tends to acumulate in bones, brain, liver and muscles, [UNSCEAR, 2000]. This radionuclide is toxic both due to its chemical properties and due to its radiological properties [Strumińska, 2016]. The radionuclide ^{210}Po and ^{210}Pb can be present in human body, due the disintegration of radionuclide ^{226}Ra, also [UNSCEAR, 2000].

Table 1.4 *- Description of the lead-210*

Description	**Numerical value**
Atomic mass, A	209,98 g/mol
Atomic number, Z	84
Half life, $T_{1/2}$	22.20 years
Belong to the radioactive series	the radioactive series ^{238}U
Daughter-nuclide	^{210}Bi
Decay mode	100% beta
Maxime Energy	76.4 Ci/g

Radium-226:

Radium-226 has a similar calcium metabolism, therefore it tends to accumulate in considerable amount in bones [J. Molinari, 1990]. Radium-226 has an important contribution to the *effective dose* received by the human body both by inhalation and by ingestion, also having a high conversion factor from activity to high dose, but also a high half-life, [Abbasi A., 2018].

Table 1.5 *- Description of the radium-226*

Description	**Numerical value**
Atomic mass, A	226,02 g/mol
Atomic number, Z	88
Half life, $T_{1/2}$	1600 years
Belong to the radioactive series	the radioactive series ^{238}U
Daughter-nuclide	^{222}Rn
Decay mode	100% alpha
Maxime Energy	0.989 Ci/g

Thorium-232:

Thorium-232 entered into the body by ingestion is absorbed and transported by the blood, preferentially accumulating in the liver and bones. This is because, in the form of Th (IV), it is close in size to Fe (III) - from the blood, for which it is easily transported into the body and deposited in the liver, [UNSCEAR, 2000]. If it enters the body by inhalation, it accumulates primarily in the lungs, liver and muscles, [Akhter P., 2007], [Adeniji A. E., 2013].

Table 1.6 *- Description of the thorium-232*

Description	Numerical value
Atomic mass, A	232.038 g/mol
Atomic number, Z	90
Half life, $T_{1/2}$	1.4 10^{10} years
Belong to the radioactive series	^{232}Th –head of series
daughter-nuclide	^{228}Ra
Decay mode	100% alpha
Maxime Energy	1.1 10^{-7} Ci/g

Uranium-238

Uranium is found in nature in oxidation state IV. This form is soluble in the blood, as a result it does not form stable compounds, the result being excretion from the body after a biological time of about four days. Due to this property of being excreted from the body, uranium is known to be nephrotoxic, thus accumulating in the kidneys [Rožmarić M., 2012]. Uranium-238 is retained also, in the body and in the skeleton. Similar concentrations have been shown in different types of bones: vertebrae and femur [UNSCEAR, 2000].

Table 1.7 *- Description of the uranium-238*

Description	Numerical value
Atomic mass, A	238,05 g/mol
Atomic number, Z	92
Half life, $T_{1/2}$	4.46 10^9 yers
Belong to the radioactive series	^{238}U - head of series
daughter-nuclide	^{234}Th
Decay mode	100% alpha
Maxime Energy	3.36 10^{-7} Ci/g

2. MATERIALS AND METHODS

In order to evaluate of the annual effective dose due to ingestion of natural rdionuclides from food and water the specific activities of the natural radionucles from these were determinated. There were determinated:

-gross alpha activity, gross beta activity and specific activities of ^{210}Po, ^{210}Pb, ^{226}Ra,^{238}U, and ^{232}Th from drinking water and mineral water,

- gross alpha activity, gross beta activity and specific activities of ^{210}Po, ^{210}Pb, ^{226}Ra,^{238}U, ^{232}Th and ^{40}K from food.

The analyzes were performed into: Ionizing Radiation Hygiene Laboratory Galati - Department of Public Health Galati, European Center of Excellence for the Environment, Faculty of Sciences and Environment - „Dunărea de Jos" University of Galați, INPOLDE International interdisciplinary network laboratories established at Dunarea de Jos University of Galati within the European funded project Romania-Ukraine-Republic of Moldova MIS ETC code 1676 and, also, the project Strategy and actions for preparing the national participation in the DANUBIUS-RI Project - acronym DANS, funded by Romanian Ministry of Research and Innovation, (the project 4/2018).

One of the ways of ensuring the quality of the results is the participation in proficiency tests. Ionizing Radiation Hygiene Laboratory Galati annually participates in such proficiency tests. One of these tests organized by Joint Research Centre - Institute for Reference Materials and Measurements, Geel, Belgia, was described by Jobbágy et al., and Sobiech-Matura et. al., [Jobbágy et al. 2014], [Sobiech-Matura et al. 2015], [Jobbágy et al. 2016].

Figure 2.1 *- The sampling area in Romania (Galati, Braila and Vrancea counties)*

The water and food samples were collected from three counties located in the South- Eastern region of Romania: Galati, Braila, and Vrancea (*Figure 2.1*). The Galati county is situated between 45°25′ and 46°10′ North latitude and 27°20′ and 28°10′ East longitude [http://statistici.insse.ro:8077/tempo-online/#/pages/tables/insse-table], Vrancea county between 45°23′ and 46°11′ North latitude and 26°23′ and 27°32′ East longitude [http://statistici.insse.ro:8077/tempo-online/#/pages/tables/insse-table] and Braila county between 44°50' and 45°20' North latitude and 21°15' and 20°00' East longitude, http://statistici.insse.ro:8077/tempo-online/#/pages/tables/insse-table

2.1. Methods to determine the radioactive level in drinking water

Measurement of the gross alpha/beta activity

The gross alpha activity and gross beta activity measurements represent the general screening of the radiological quality of drinking water.The methods used for measurement of the gross alpha and gross beta activity are in accordance with ISO 9696 and ISO 9697.

Firstly, the dry residue content was determined for all samples. The knowledge of this value is very helpful to calculate the volume of samples required to produce a sufficient residue for alpha and beta measurement. Secondly, the sample was evaporated, converted to sulphate, and calcined. After that, the residue uniformly distributed onto stainless planchet was measured. The measurements were performed using the low background MPC-2000-DP and MPC-900-DP (Protean Instruments Corporation) counting system with a ZnS dual phosphor detector (zinc sulphide and plastic) calibrated at Horia Hulubei National Institute of R&D in Physics and Nuclear Engineering (IFIN-HH), Magurele. ^{241}Am point reference source (serial:2830 LMRI France) and ^{90}Sr/Y point reference source (serial:9891) were used for current calibration in Ionizing Radiation Laboratory. For this instrument through alpha mode measurements and beta mode measurements, a ratio of 1:10 was observed of residue weights for alpha measurements and beta measurements transferred to a stainless steel planchet [Pintilie V., 2016a]. The time for measurements was 100 min.

The activity concentrations of the gross alpha and gross beta activity were calculated using the formula :

$$\Lambda_{\propto\, or\, \beta} = \frac{(R_{sample} - R_0) \times TDS}{\varepsilon \times m} \quad (Bq \cdot L^{-1}) \qquad (2.1)$$

where $\Lambda_{\alpha/\beta}$ is the activity concentration of the gross alpha/beta activity of the drinking water sample (Bq L^{-1}), R_{sample} is the rate of alpha/beta measurement for the sample (counts·s^{-1}), R_0 is the rate of alpha/beta measurement for background (counts·s^{-1}), TDS is the concentration of the total dissolved solids of the sample (g L^{-1}), *m* is the weight of the residue transferred to a stainless steel planchet for measurements (g) and *ε* is the efficiency of the detector.

Determination of ^{210}Po and ^{210}Pb concentration

In the present study specivic activities of ^{210}Po și ^{210}Pb from water were performed by radiochemical separation through SR resin. Eichrom's Sr Resin contains 4,4′(5′)-di-t-butylcyclohexano 18-crown-6 (crown ether) in 1-octanol. The method was learned by "*Training in radiochemistry measurements for practitioners from countries eligible under the JRC Enlargement & Integration Policy*" (the traning was performed at the "Jožef Stefan" - Ljublijana – Slovenia, 10-21 nov. 2014), and published: [Benedik L., 2009], [Rožmarić M., 2012], [Benedik L., 2012].

The principle of ^{210}Po and ^{210}Pb isotope determination consists of: concentration of the sample by co-precipitation of the polonium and lead, dissolving the precipitate, resulting the solution with active components (^{210}Po, ^{210}Pb), separating the lead from polonium on the SR resin - when eluted from polonium, respectively eluted of lead, *Figure 2.3.*

Figure 2.3 – *Concentrating of ^{210}Po, ^{210}Pb by coprecipitation, separated through the columns with SR-resin and purification*

The specific activity of ^{210}Po is calculated as:

$$\Lambda_{210Po} = \frac{(R_{\text{sample}} - R_0)}{V \times \varepsilon \times \eta} \quad (Bq\ L^{-1}) \qquad (2.14)$$

where: Λ_{210Po} is the activity concentration of the sample (Bq L^{-1}), R_{sample} is the rate of measurement for the sample (counts·s^{-1}), R_0 is the rate of measurement for background (counts·s^{-1}), V is the volume of the sample (L) and η is the chemical recovery of radiochemical separation, ε – is the efficiency of the detector (imp/sec/Bq).

The specific activity of ^{210}Pb is calculated as:

$$\Lambda_{210Pb} = \frac{(R_{210Bi} - R_0)}{\varepsilon \times \eta \times \left(1 - e^{\frac{ln2 \times \Delta t}{T_{\frac{1}{2}}}}\right) \times V} \quad \left(\frac{Bq}{l}\right) \qquad (2.16)$$

where: Λ_{210Pb} – the activity concentration of the sample ^{210}Pb, (Bq L^{-1}); R_{210Bi} – the rate of measurement for precipitate (counts·s^{-1}), R_0 – the rate of measurement for background (counts·s^{-1}), V – the volume of the sample (L), ε – is the efficiency of the detector (imp/sec/Bq), η – randament de separare radiochimica (%), Δt – the time between separation of ^{210}Pb and measurement of ^{210}Bi (days); $T_{1/2}$ – half life ^{210}Bi (days).

Determination of ^{226}Ra concentration

The water samples were stored 30 days in a sealed system before the determination of the activity of ^{226}Ra, *Figure 2.5*. At the end of this period, enough ^{222}Rn was built up from ^{226}Ra water samples.

Figure 2.5 *– The sample sealed and stored for 30 days*

After this period, the bubbling flask was connected to the module SARAD RTM 1688-2, without being opened, using Hoffman clamps, *Figure 2.6*. After the connection of the bubbling flask to the measurement system, the Hoffman's clamp was opened in such a way that the water did not come into contact with the air outside.

Figure 2.6 *- SARAD RTM-1688-2 measuring system for ^{226}Ra/^{222}Rn in water [Pintilie V., 2016b]*

The acquired measurement data are downloaded to the serial interface. The software calculates the activity concentration of ^{226}Ra (Bq $^{L-}$1) using the equation 2.28:

$$\Lambda_{226Ra/222Rn} = \frac{1}{V_{sample}} \times C_{aer} \times \left[k \times V_{probă} + V_{aer}\right] \; (Bq \cdot \mathrm{L}^{-1}) \qquad (2.28)$$

where: $\Lambda_{226Ra./222Rn}$ – the activity concentration of ^{226}Ra/^{222}Rn (Bq·L^{-1}), V_{sample} – the volume of the sample (L), C_{air} –the value indicated by the measuring system (Bq·L^{-1}),V_{air} – the volume of the air from the measuring system (L), k – the temperature correction factor.

2.2. Methods to determine the radioactive level in food samples

The general scheme used for the primary processing of the food samples and the distribution of the aliquot residue/ash parts, from the same sample can be found in *Figure 2.8.* The values of these quantities were used to determine the natural radionuclides from foodstuffs.

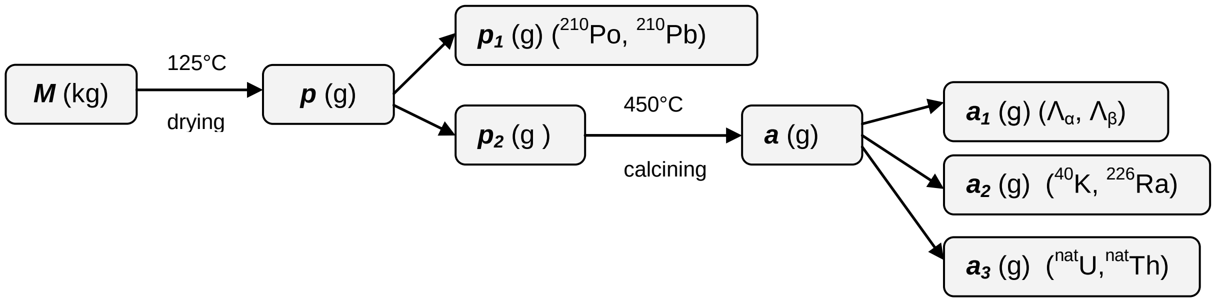


Figure 2.8 *- General scheme of the foodstuffs sample primary processing and the distribution of the aliquot residue/ash parts in order to determine the gross alpha, gross beta and ^{210}Po, ^{210}Pb, ^{238}U, ^{232}Th, ^{226}Ra and ^{40}K activity [Pintilie V., 2018b]*

Measurement of the gross alpha/beta activity

The following equation was used to calculate the gross alpha activity and gross beta activity:

$$\Lambda_{\propto\ or\ \beta} = \frac{(R_{probă} - R_0) \times p \times a}{\varepsilon \times M \times p_2 \times a_1} \quad (Bq \cdot kg^{-1}) \qquad (2.29)$$

where $\Lambda_{\alpha\ or\ \beta}$ -is the gross alpha activity or gross beta activity of the sample ($Bq \cdot kg^{-1}$), R_{sample} - is the rate of measurement of the ash transferred onto the planchet ($counts \cdot s^{-1}$), R_0 is the rate of alpha, beta background (count s^{-1}), respectively. *a* - is the total mass of ash resulting from the calcinations at 450˚C (g), p – is the total mass of the residue resulting from drying at 125˚C ε – is the efficiency of the detector (imp/sec/Bq); p_2 - an aliquot residue part which is calcined (g); a_1 - an aliquot ash part measured (g)

Determination of ^{210}Po and ^{210}Pb concentration

An aliquot part (p_1 grams, *Figure 2.8*) from residue was mineralised It was used as the solution for self deposition (90 °C, 4 h mixing) onto nickel disc. After that the gross alpha activity of the nickel disc was measured. Using the equation 2.33 the concentration of ^{210}Po is calculated:

$$C_{210Po} = \frac{(R_{disc} - R_0) \times p}{\varepsilon \times \eta \times M \times p_1} \quad (Bq \cdot kg^{-1}) \qquad (2.33)$$

where C_{210Po} is the concentration of ^{210}Po ($Bq \cdot kg^{-1}$), R_{disc} is the rate of measurement of self deposited nickel disc ($counts \cdot s^{-1}$), R_0 is the rate of measurement for background ($counts \cdot s^{-1}$), ε is efficiency of the detector of MPC-900-DP system and η is radiochemical yield. After 3 months, it was used the rest of the solution for second self deposition onto nickel disc. The radionuclide ^{210}Pb was determined from the solution generating ^{210}Po, which was separated in the second self deposition. The concentration of ^{210}Pb was calculated with the following equation:

$$C_{210Pb} = \frac{(R_{disc} - R_0) \times p}{\varepsilon \times \eta \times M \times p_1 \times 0.37} \quad (Bq \cdot kg^{-1}) \qquad (2.34)$$

where C_{210Pb} is the concentration of ^{210}Pb ($Bq \cdot kg^{-1}$), R_{disc} is the rate of measurement of the second self deposited nickel disc ($counts \cdot s^{-1}$), 0.37 is the correction factor for secular equilibrium, R_0 is the rate of measurement for background ($counts \cdot s^{-1}$), ε is efficiency of the detector— MPC-900-DP system, η is radiochemical yield.

Determination of ^{nat}U and ^{nat}Th concentration

An aliquot part of ash (a_3 g), obtained as shown in *Figure 2.8*, was subjected to mineralization through acidic extraction and, after that, ^{nat}U and ^{nat}Th were separated and purified. In the *Figure 2.11*, the specified reagents volumes were taken into consideration for approx. 5 g ash and approx. 10 mL Dowex resin.

The succession of the reagents for the separation U and Th from the acidic extract through the column with prepared Dowex resin in Cl^- form is shown schematically in *Figure 2.11*.

The method does not allow the addition of the internal radioactive tracer and the determination of the radiochemical yield concomitant with the determinations in the sample. Therefore, the separation efficiency is done separately, on samples fortified with certified reference materials.

The values obtained in this procedure for the radiochemical yield are 61% for ^{nat}U and 59% for ^{nat}Th. Taking into consideration the natural isotopic abundance of ^{238}U (99.27%) and ^{232}Th (100%) from ^{nat}U and ^{nat}Th, respectively, the specific activities of ^{238}U and ^{232}Th were calculated.

Determination of ^{40}K and ^{226}Ra concentration by gammaspectroscopy

The concentrations of ^{40}K and ^{226}Ra were determined using gamma-spectrometry as described in [Pintilie V., 2017]. For the measurements a NaI(Tl)-detector coupled to a multichannel analyzer and Maestro-32 software were used. The gamma spectrometer was calibrated using a mixed source SEG 8-471 (^{60}Co, ^{137}Cs, ^{241}Am), produced at IFIN-HH. The concentration of ^{40}K was determined using its gammaray peak of 1460 keV, isotopic abundance of 0.0117% and ε=(7.40 ± 0.44)x10^{-3}, $counts \cdot s^{-1}$ Bq^{-1}. The concentration of ^{226}Ra was determined using its progeny, ^{214}Bi, with peak energy of 1120 keV, isotopic abundance of 15% and ε =(10.00 ± 0.54)x10^{-3} $counts \cdot s^{-1}$ Bq^{-1}. The samples were sealed and stored for 4 weeks to reach the equilibrium and then counted for 7200 s. The measurement geometry was the same as the calibration geometry, *Figure 2.12*

Figure 2.12 *- The radioactive source used for calibration of the spectrometer and the measuring sample - identical geometry*

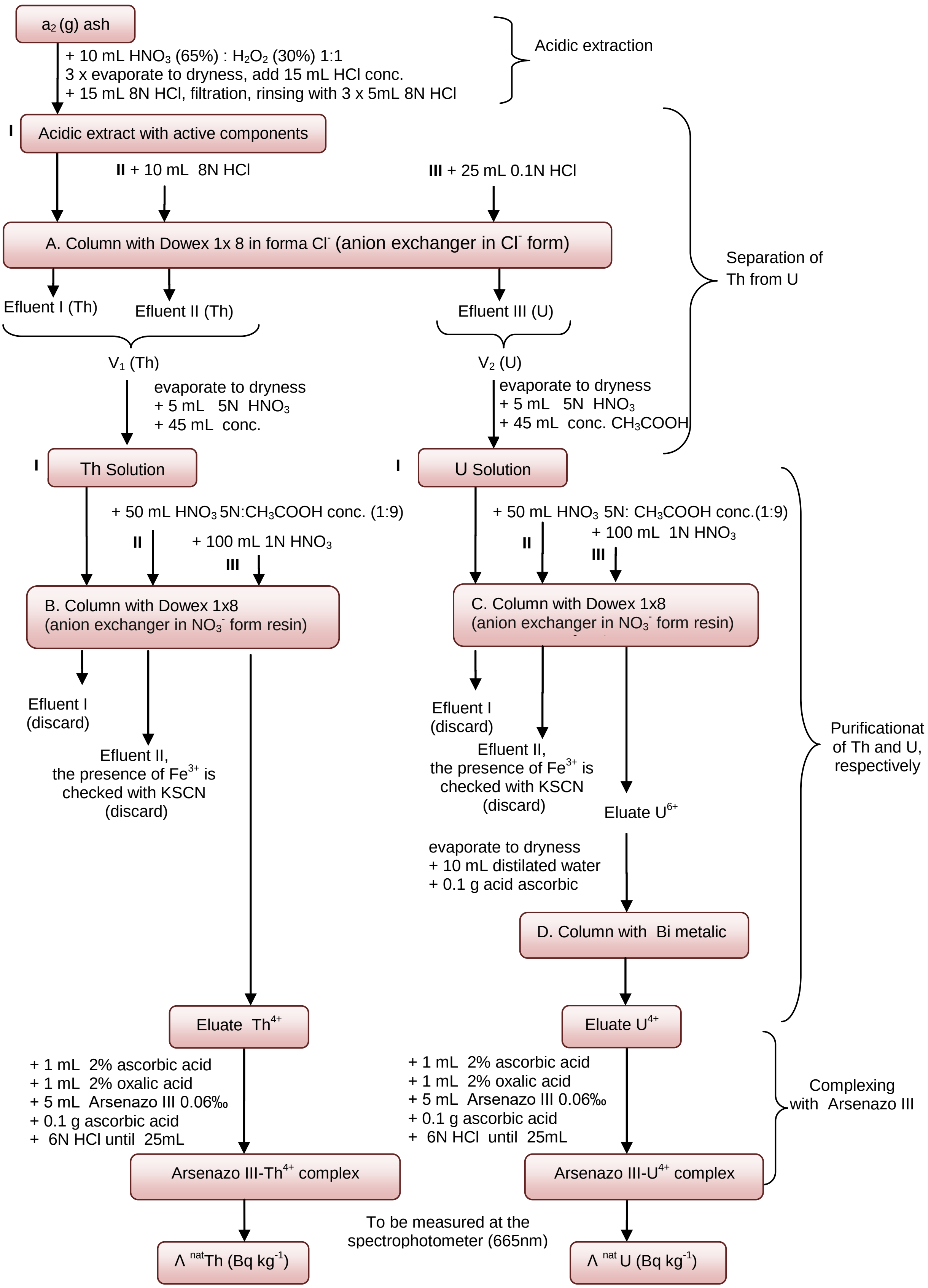


Figure 2.11 *- Scheme of radiochemical procedure of* ^{nat}*U and* ^{nat}*Th from foodstuffs sample*

Assessment the effective dose due to ingestion of radionuclides from foodstuff consumption

Using the conversion factors published by the Protection and Safety of Radiation Sources: *International Atomic Energy Agency* [IAEA, 2017] and the annual foodstuffs consumption rate for Romania [http://statistici.insse.ro. Accessed 30 Nov 2017], the annual effective dose, is calculated [Ene A., 2017], [Pintilie V., 2018a], [Pintilie V., 2018b], using the following formula:

$$D_{ef} = \sum(\Lambda_X \times R \times CF)\ (Sv \cdot year^{-1}) \qquad (2.48)$$

where Λ_X is the concentration of the radionuclide X (X=^{210}Po, ^{210}Pb, ^{238}U, ^{232}Th, ^{40}K, ^{226}Ra) (Bq kg^{-1}), R is the annual foodstuffs consumption (kg year^{-1}), and CF is effective dose conversion factor for adults (Sv·Bq^{-1}). For the calculations the following dose conversion factors for adults were used: 1.2x10^{-6} Sv·Bq^{-1} for ^{210}Po, 6.9x10^{-7} Sv·Bq^{-1} for ^{210}Pb, 4.5x10^{-8} Sv·Bq^{-1} for ^{238}U, 2.3x10^{-7} Sv·Bq^{-1} for ^{232}Th, 6.9x10^{-9} Sv·Bq^{-1} for ^{40}K, and 2.8x10^{-7} Sv·Bq^{-1} for ^{226}Ra [IAEA, 2014].

Assessment of the annual effective dose due to ingestion of radionuclides from water consumption

The quantification of the ionizing radiation effects on the human body due to the consumption of drinking water is performed by calculating the physical magnitude called the annual effective dose - expressed in Sv year^{-1}, defined in Council Directive 2013/51/EURATOM as *the committed effective dose for one year of ingestion resulting from all the radionuclides whose presence has been detected in a supply of water intended for human consumption, of natural and artificial origin, but excluding tritium, potassium-40, radon and short-lived radon decay products*.

The European legislation [Directive 51/2013] transposed into Romanian legislation [L301/2015] recommends that this value parameter should not exceed 0.1 mSv year^{-1}.

In the scientific literature, different modes of calculation were identified for evaluate of the annual effective dose due to ingestion of radionuclides from water consumption:

- based on the determined concentration of investigated radionuclides calculated [Jia G., 2009], [Rajashekara K. M., 2011], [Walsh M., 2014], [Altıkulaç A., 2015], [Rožmarić M., 2012], [Pintilie V., 2016a], [Pintilie V., 2016b], [Farthabadi N., 2019];
- using the gross alpha activity [Fenandez J. F., 1992], [Kobya Y., 2015], [Abbasi A., 2017], [Turhan Ș., 2013] [Sajo-Bohus L., 1996];
- using the gross alpha and gross beta activity [Gorur F. K., 2014] , [Korkmaz M. E., 2016], [Ogundare F. O., 2015], [Pintilie V., 2016a];
- assuming the gross alpha activity to alpha radionuclides (e.g.: ^{210}Po, ^{226}Ra, ^{232}Th, ^{238}U), and the gross beta activity to the beta radionuclides (e.g.: ^{228}Ra, ^{210}Pb) [Gorur F. K., 2014], [Akbulut S., 2015], [Pintilie V., 2016a].

The relevant advantages and disadvantages regarding the calculation algorithms for the annual effective dose are shown in the *Table 3.5*

In the present study, the calculation methods of the annual effective dose due to the ingestion of natural radionuclides through of drinking water were analysed, exemplified and discussed. This was done on the basis of the *measured* radioactivity parameters, on the one hand, and on the basis of the *assumed* radioactivity parameters, on the other hand, using the calculation rationale found in the domain-specific literature.

Table 3.5 - *The advantages and disadvantages - the calculation algorithms for the annual effective dose*

No	The methods	The advantages	The disadvantages
1	The assessment of the total effective reference dose using the measured concentrations of the investigated radionuclides	- the accuracy of the result is high, - provides information on the contribution of radionuclides determined at the dose.	- high response time, -radiochemical (reagent-time-consuming) determinations of radionuclides with activity-to-high conversion factor is required
2	The assessment of the total effective reference dose using the assumed concentrations based on gross alpha and beta activities	- fast response time - cover the maximum risk - only radiometric determinations are required: alpha and beta global activity - covers the maximum risk due to radiation effects	- the accuracy of the result is low -the ^{40}K contribution from global beta activity is not subtracted
3	The assessment of the total effective reference dose using the gross alpha activity	- fast response time -only one type of determination is required: global alpha activity	- the accuracy of the result is low -not provide information on the contribution of radionuclides to the dose
4	The assessment of the effective reference dose by associating the gross alpha activity and the gross beta activity with the alpha emitters (ex ^{210}Po, ^{226}Ra, ^{232}Th, ^{238}U), and with the beta emitters, respectively (^{228}Ra, ^{210}Pb)	- fast response time -only radiometric determinations are required: alpha and beta global activity	- the accuracy of the result is low -the ^{40}K contribution from global beta activity is not subtracted

Depending on the laboratory (both from a technical and from a specialized human resource point of view), on the available data, on the response time available, on the aim pursued, the most adequate method of D_{ef} assessment is selected, each of the above-mentioned methods providing valuable information on quantifying the exposure of the population to ionizing radiation by means of drinking water consumption.

Detriment due to ionising radiations

Detriment is used to evaluate of the negative effects on human health due to ionizing radiation. Lifetime risk assessment was carried out using the formula:

$$LR = D_{ef} \times L \times RF \qquad (2.50)$$

where: LR is lifetime cancer risk, D_{ef} is the annual effective dose ($Sv \cdot year^{-1}$), L is the duration of life (year) and RF is the risk factor for fatal cancers for the whole population $5{,}5 \times 10^{-2}$ Sv^{-1}, [ICRP, Publication 103].

3. EXPERIMENTAL RESULTS AND DISCUTIONS

This is the first detailed study to evaluate the exposure of the population to ionizing radiation due to the ingestion of natural radionuclides (^{210}Po, ^{210}Pb, ^{226}Ra, ^{nat}U, ^{40}K) through the consumption of drinking water, mineral water and food in counties: Galați, Brăila and Vrancea located in the South- Eastern region of Romania. It is also the first detailed study on the determination of the natural radioactive content from: drinking water, mineral water and food such as: bread, pasteurized milk, meat, milk powder for children in the age group 0-12 months, daily menu for children in the category of age 2-7 years.

In order to evaluate the annual effective dose due to the ingestion of natural radionuclides through the consumption of drinking water and food, the following research directions were carried out within this thesis:

• determination of radioactivity parameters in drinking water - samples were collected from public drinking water supply networks in Galați, Brăila and Vrancea counties;

• determination of the radioactivity parameters in mineral water - the samples were collected from supermarkets located in Galați county;

• determining the parameters of radioactivity in milk powder for children in the age category 0-12 months - the samples were collected from pharmacies located in Galați county;

• determination of radioactivity parameters in the daily menu - the samples were collected from kindergartens with extended program located in Galați, Brăila and Vrancea counties;

• determination of the parameters of radioactivity in bread - the samples were collected from supermarkets located in Galați, Brăila and Vrancea counties;

• determining the parameters of radioactivity in pasteurized milk - the samples were collected from supermarkets located in Galați, Brăila and Vrancea counties;

• determination of the radioactivity parameters in the meat - the samples were collected from supermarkets located in Galați, Brăila and Vrancea counties;

Based on the radioactivity parameters determined, for the population in the region of Galați, Brăila and Vrancea counties, the annual effective dose due to the consumption of drinking water, mineral water and food was evaluated.

In order to evaluate the annual effective dose due to drinking water consumption, two evaluation algorithms were used, described in Chapter 3, paragraph 3.1 (from thesis), namely: the first based on the gross alpha and gross beta activity and the second, based on the concentration values. of radionuclides ^{210}Po, ^{210}Pb, ^{226}R and ^{nat}U. Discussions on the advantages and disadvantages of annual effective dose calculation algorithms are presented in *Chapter 3*.

Using the primary parameters of radioactivity in drinking water: alpha and global beta activity, maps were made regarding their spatial distribution, for Galați and Vrancea counties, respectively maps regarding the spatial distribution of the annual effective dose due to drinking water consumption.

To calculate the annual effective dose due to food consumption, a single calculation algorithm was used, namely that based on the determined concentrations of the radionuclides of ^{210}Po, ^{210}Pb, ^{226}Ra, ^{nat}U and ^{40}K.

Using the values of the effective annual dose thus determined, the *detriment* due to the ionizing radiation brought to the health of the population by the consumption of drinking water and food was evaluated and compared with the *maximum detriment* evaluated for the population of Galaţi, Brăila and Vrancea counties, corresponding to the same study period.

Another research direction of this thesis was the evaluation of the *annual effective dose* due to the inhalation of radon from the air of public buildings and dwellings. For this, radon from schools and dwellings was measured, after which the corresponding effective annual dose was evaluated.

3.1. Assessment to the annual effective dose due to drinking and mineral water consumption

The assessment of the annual effective reference dose using the gross alpha activity

Based on the gross alpha and gross beta activity were evaluated:
- the annual effective dose due to drinking water consumption from Galați county, during from 2014 to 2017;
- the annual effective dose due to drinking water consumption from Vrancea county, during from 2015 to 2018;
- the annual effective dose due to drinking water consumption from Brăila county, during from 2014 to 2018.

During the period from 2014 to 2017, the gross alpha and the gross beta activity from drinking water distributed from Galati county were determined. The values of these parameters were transposed into spatial distribution maps of the radioactive content in the drinking water.

The analises of the maps in *Figures 3.1, 3.2, 3.3, 3.4* (Chapter 3, subchapter 3.2 from extended thesis), representing the value domains of the gross alpha and gross beta activity, corresponding to the samples collected from Galați county, during the period 2014-2017 (only the *Figure 3.1* and *Figure 3.2* are shown in this abstract thesis), it was observed a dynamic of the changes in concentration more pronounced in the first two years of the studied period and weaker in the last two years. This is due to the fact that, during the course of the study, many of the drinking water supplies, initially under the care of the territorial administrative units, passed into the ownership of the main producer of drinking water in Galați county, intervening on the sources used (drilling modification - in some cases, improvement, modernization) - remaining in the last two years owned by the same manufacturer.

By superimposing the map of the gross alpha and beta concentrations with the map of the relief of Galați county, it is observed that, in general, the dynamics of changes regarding the gross alpha and gross beta activity of drinking water depend more on the geological structure of the area from which these waters originate and less on the technological processes for the production of drinking water. Otherwise, there should have been a constant concentration of radioactivity parameters (gross alpha and gross beta activity) over the entire range of drinking water sampling areas in Galați county.

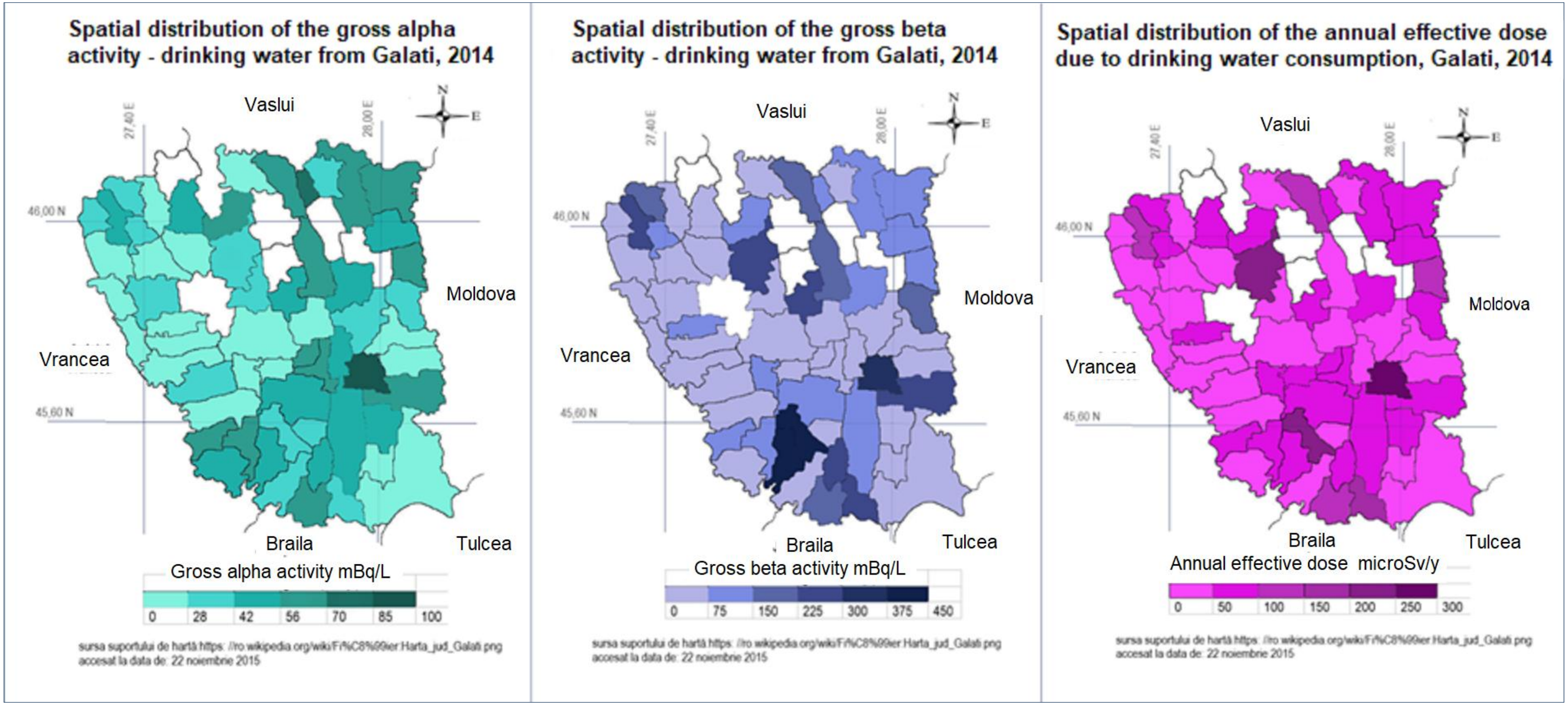


Figure 3.1 *- Spatial distribution of the gross alpha and beta activity in drinking water, and the annual effective dose due to drinking water consumption, in Galați county, 2014 (white areas-did not have drinking water facilities during the study period)*

The multivariate analysis of the radioactivity parameters determined in the drinking water collected from Galați county, in 2014, leads to the dendrogram in *Figure 4.1*, from which it is observed that a high similarity of 87-98% (the first, and second clusters) is presented by the samples collected from similar geomorphological units in terms of their structure. Thus, a similarity of the concentration range, higher, of the gross alpha activity, is observed in the case of the samples collected from the communes located on the Tecuci Plain, Covurlui Plain and the Lower Siret Plain. These geomorphological units of Galati County are located above the North-Dobrogean Orogen, therefore they have a similar sedimentary foundation and cover. The similarity, in terms of composition, of these three geomorphological units (Tecuci Plain, Covurlui Plain and the Lower Siret Plain) of Galaţi county, may explain the similarity of the radioactive content of the water samples collected from them.

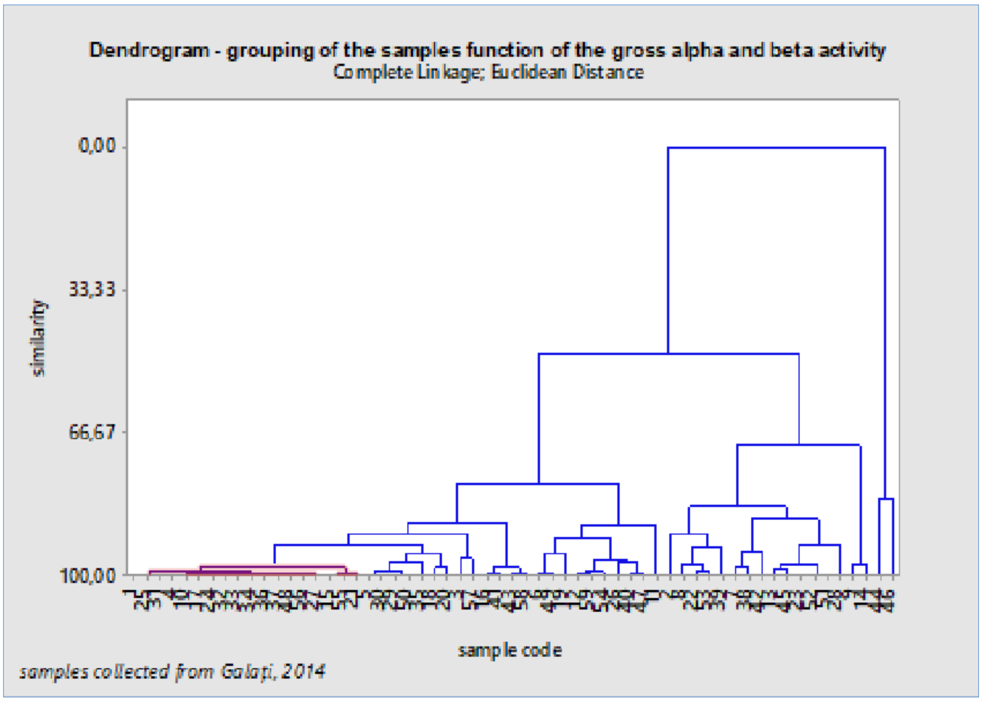


Figure 4.1 *– Dendrogram – gross alpha and beta activity for drinking water samples collected from Galati county, 2014*

Similar observations are derived from the dendrograms corresponding to the samples analyzed from Galați county in the period 2015-2017.

During the period from 2014 to 2017, from the point of view of the radioactivity parameters of the drinking water, in Galați county, the values determined for gross alpha, gross beta are range from 6 to 80 mBq L^{-1} , and from 25 to 435 mBq L^{-1}, respectively. The average values of the gross alpha and gross beta activity are 35 ± 7 mBq L^{-1} , respectively 78 ± 16 mBq L^{-1} - representing 35%, and 7.8%, respectively, from the maximum concentrations allowed for these radioactivity parameters. The gross beta activity in these samples is higher than the gross alpha activity. For all the analyzed samples from Galați county, during 2014-2017, there were no exceedances of the maximum allowed concentrations of these radioactivity parameters.

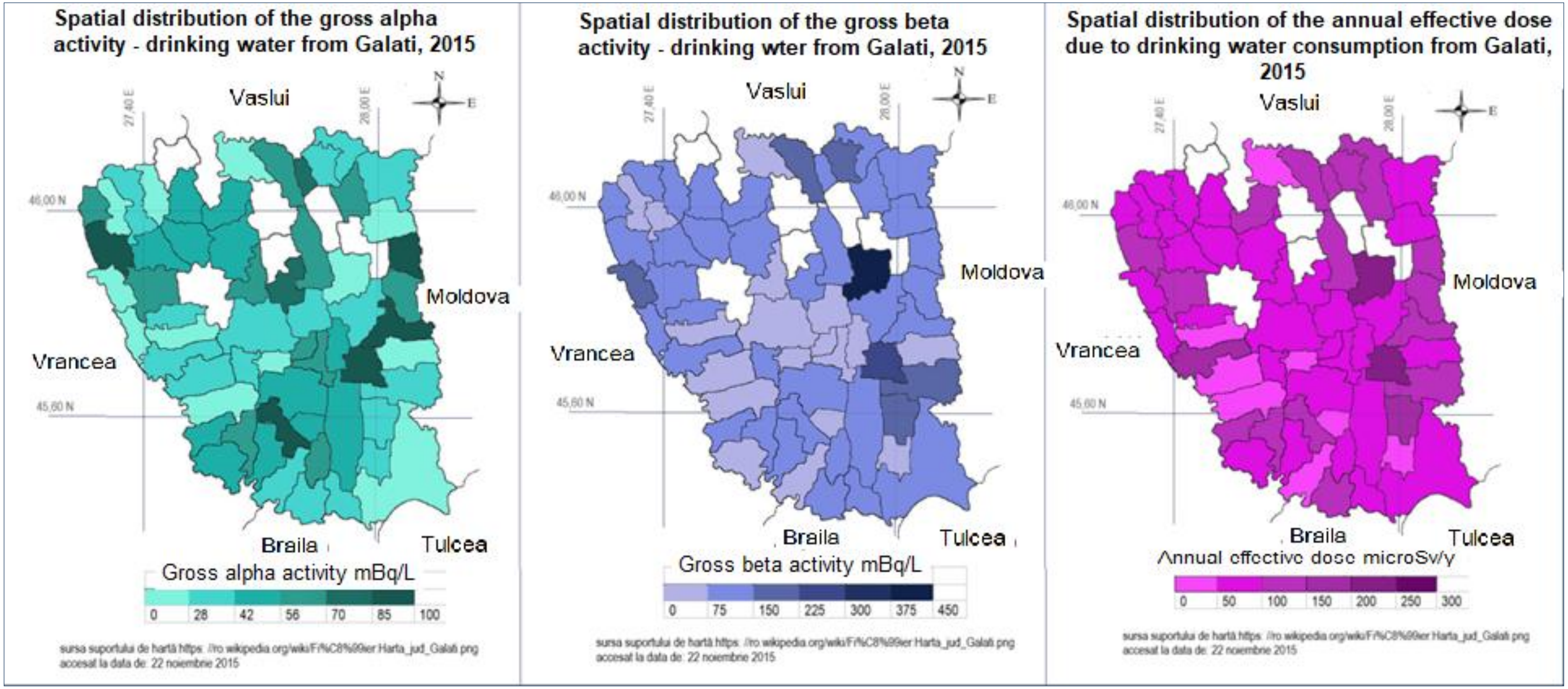


***Figure 3.2** – Spatial distribution of the gross alpha, gross beta, and annual effective dose due to drinking water consumption collected from Galati county, 2015, (the white areas - did not have drinking water distribution facilities during the study)*

The variation of the radioactivity parameters corresponding to these samples collected from Galați county, during from 2014 to 2017, are shown in *Figure 4.2.*

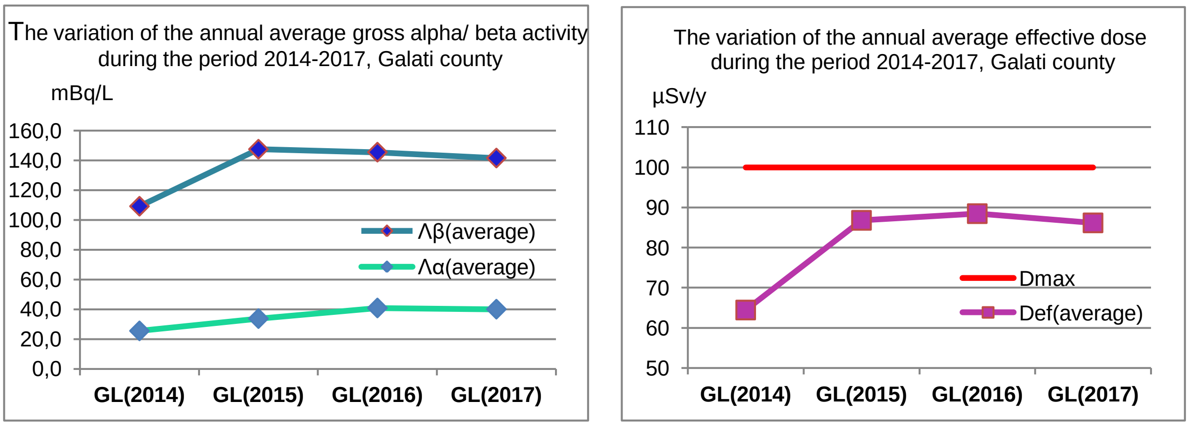


***Figure 4.2** - The variation of the annual average gross alpha/beta activity, and annual average effective dose due to drinking water consumption, during the period 2014-2017, Galati county*

The water from the Danube river, whose characteristics are described in the scientific literature [Iticescu C., 2019], [Banescu A., 2018], [Apetrei C., 2019], [Popa P., 2018], [Iticescu C., 2013] is used as the source of water for Galati and Braila counties, therefore it was also determined the natural radioactive level from surface water, namely Danube river. This research direction was conducted within the framework of the project titled *Strategy and actions for preparing the national participation in the DANUBIUS-RI Project*—acronym DANS, funded by Romanian Ministry of Research and Innovation, (the project4/2018).

The sampling and the determinations were carried out in two steps: on June and September 2018. The values of the radioactivity parameters are exemplified in *Figure 3.14* for first step,

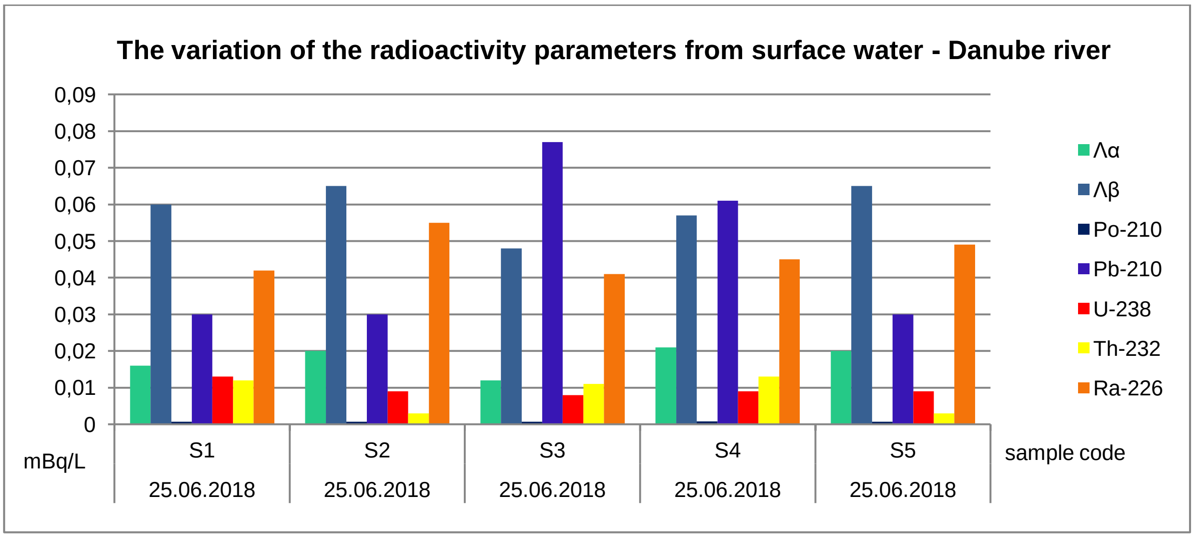


***Figure 3.14** – The variation of the radioactivity parameters (gross alpha, gross beta, concentration of ^{210}Po, ^{210}Pb, ^{238}U, ^{232}Th, ^{226}Ra) from surface water – Danube*

For the first step: radioactivity parameters determined are: 17.8 ± 5.3 mBq L^{-1} - for the gross alpha activity; 59.0 ± 14.5 mBq L^{-1} - for gross beta activity, 0.7 ± 0.2 mBq L^{-1} - for ^{210}Po concentration; 45.6 ± 13.6 mBq L^{-1} - for ^{210}Pb concentration; 9.6 ± 2.4 mBq L^{-1} - for the concentration of ^{238}U; 8.4 ± 2.1 mBq L^{-1} - for the concentration of ^{232}Th; 46.4 ± 11.6 mBq L^{-1} - for the concentration of ^{226}Ra.

There were no major changes in the concentration of natural radionuclides (^{210}Po, ^{210}Pb, ^{238}U, ^{232}Th, ^{226}Ra) in the surface water of the Danube river in the Galati segment, depending on the sampling time of the sample, sign that the migration of natural radionuclides into water depends more less on the sampling time and depends more on the radioactive and chemical content of the geological layers crossed.

Although for the concentrations of natural radionuclides in surface waters there are no maximum permitted limits in the legislation, comparing the obtained results, for the radioactivity parameters determined, with the maximum allowed concentrations for drinking water, it is observed that these are not exceeded except for the concentration of ^{210}Pb (CMA_{Pb-210} = 0.025 Bq L^{-1}). This radionuclide comes from the natural radioactive series of uranium-238, present in all natural components of the environment.

The annual effective dose due to the drinking water consumption, in Galați county, between 2014-2017, was evaluated, using the formula (3.7) - (*Chapter 3, sub-chapter 3.1*), at the average value of 81.5 ± 15.6 μSv $year^{-1}$. The variation of *annual effective dose* in Galați county, between 2014-2017, has the same tendency corresponding to the variation of the gross alpha and beta activity, *Figure 4.2.*

The risk assessment due to the annual effective dose received by the population of Galaţi county through the consumption of drinking water, for the period 2014-2017, led, for adults, to the value of 34.9×10^{-5}, compared to 42.8×10^{-5} - value of the risk, calculated if the annual effectiv dose reaches the value of 100 μSv $year^{-1}$, representing the maximum allowed limit, according to Law 301/2015 and the recommended value not to be exceeded - according to European Directive 51/2013. For to evaluate this risk, the formula (3.5) - (Chapter 3, subchapter 3.1) was used. For the average human lifespan, the value of 75.20 years was used, representing the average lifespan in Galați county, from 2014-2017, [http://statistici.insse.ro:8077/tempo-online, accessed on 12.02.2018], (see Table 4.7) and for the nominal risk coefficient adjusted to the detriment of cancer and genetic effects (Sv^{-1}) the present value of 5.7×10^{-2} Sv^{-1} was used [ICRP, Publication 103].

For the population from Vrancea county, in the period 2015-2018, based on the determination of the primary parameters of radioactivity, gross alpha activity and gross beta, the annual effective dose due to drinking water consumption was evaluated. The data obtained are transposed into the spatial distribution maps of the gross alpha activity, the gross beta, as well as the annual effective dose due to drinking water consumption - shown in Figures 3.5, 3.6, 3.7 and 3.8 (only the *Figure 3.7* and *Figure 3.8* are shown in this abstract thesis),

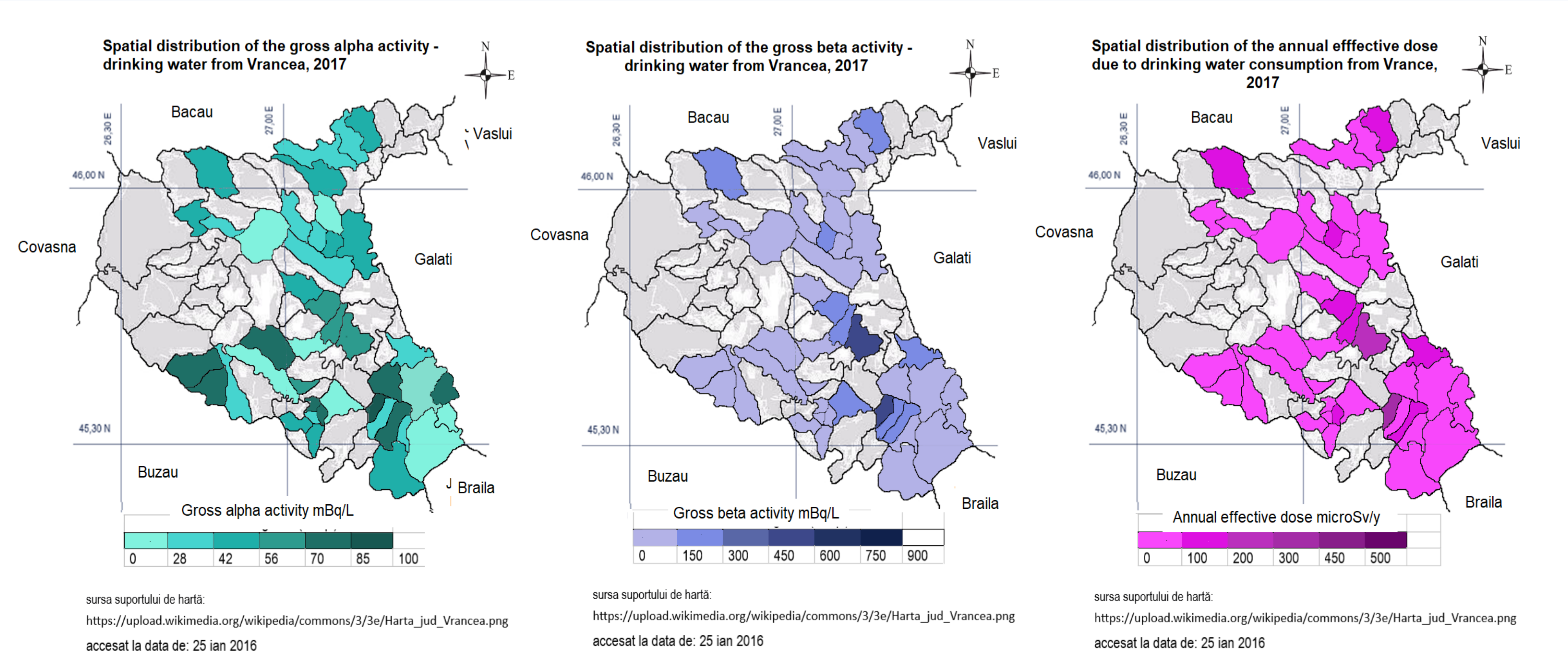


Figure 3.7 *- Spatial distribution of the gross alpha, gross beta, and annual effective dose due to drinking water consumption collected from Vrancea county, 2017, (the grey areas - there did not collect samples)*

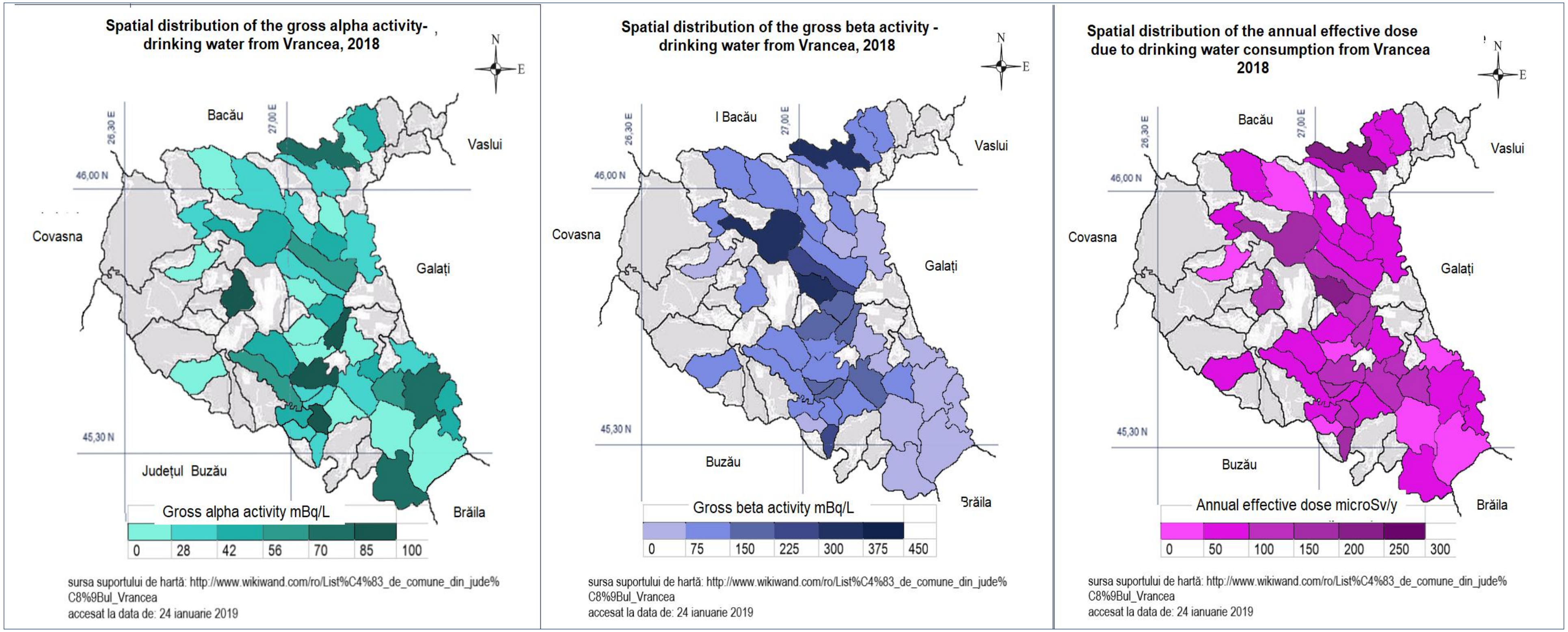


***Figure 3.8** - Spatial distribution of the gross alpha, gross beta, and annual effective dose due to drinking water consumption collected from Vrancea county, 2018, (the grey areas - there did not collect samples)*

Applying the multivariate analysis technique by grouping drinking water samples according to the values of the gross alpha and gross beta activities, for drinking water samples colected from Vrancea during 2015, the dendrogram from *Figure 4.3* resulted:

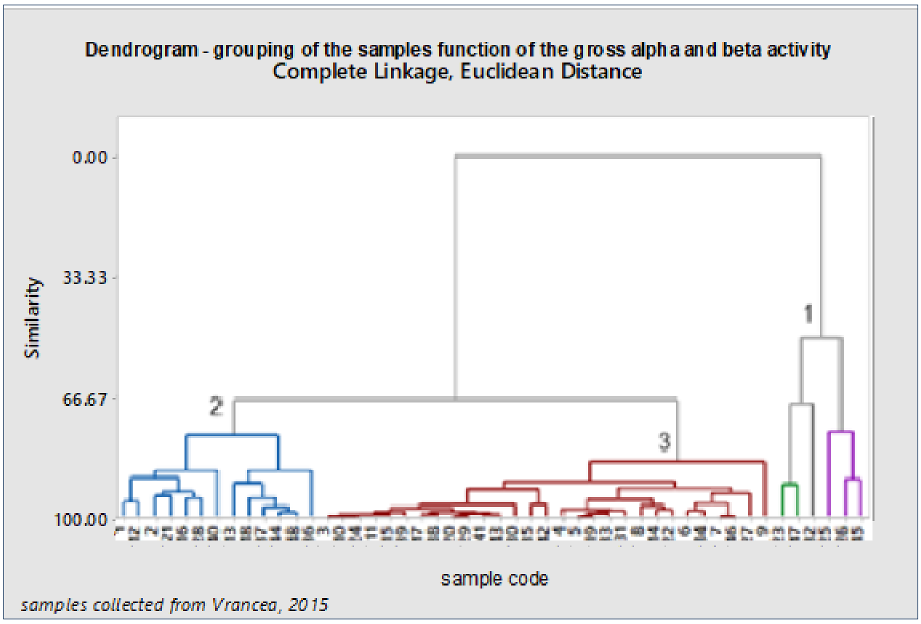


Figure 4.3 *- Dendrogram – Grouping of the samples function of the gross alpha and beta activity (drinking water samples collected from Vrancea couny, 2015)*

Cluster no. 3, shown in *Figure 4.3* with the red color, groups the samples that have a similarity, in terms of global radioactive content, of 90%. These were placed, highlighted in green color, on the geographical map of Vrancea county, corresponding to the sampling site, for a better visualization of the link between the radioactive content and the location on geomorphological units.

Analyzing the geographical location of the sampling places for drinking water samples belonging to the cluster no. 3, it is observed that this outlines the Lower Siret Plain, more precisely the High Plain within it, *Figure 4.4*.

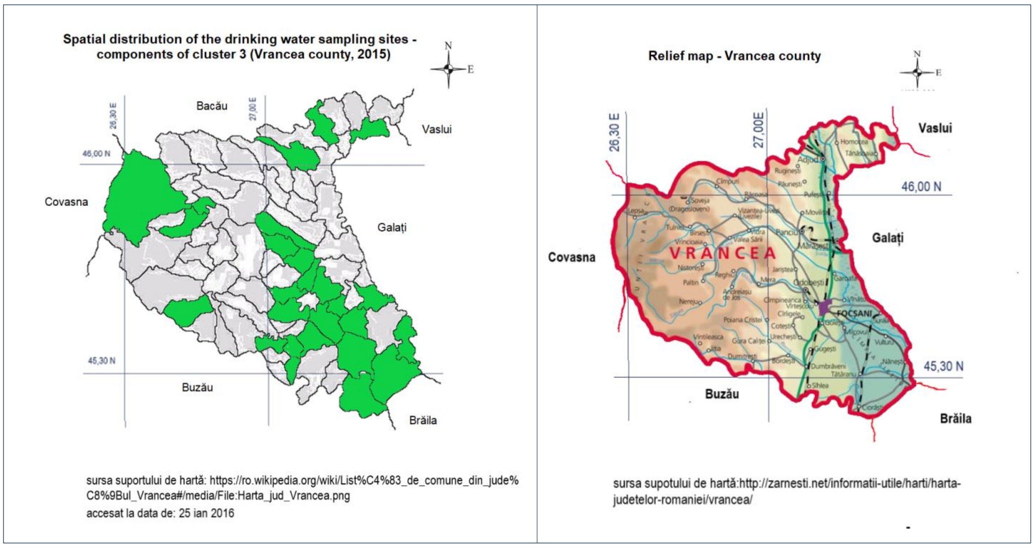
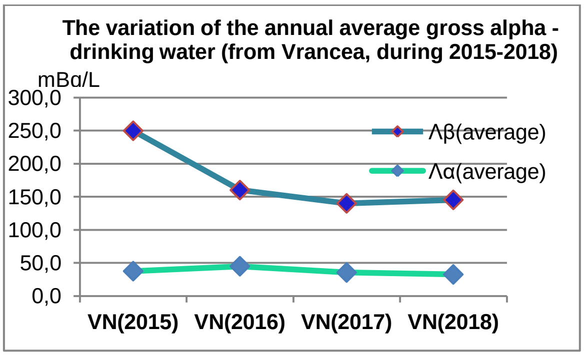


Figure 4.4 – *Spatial distribution of the area's collected samples – components of the cluster no. 3, comparative with the relief map's Vrancea county*

The similarity between the component samples of the cluster no. 3 can be put on account of the geological similarity of the sampling sites.

During four years of study, in Vrancea County, the annual average value for global alpha activity varies in a much narrower area than the annual average of global beta activity values, so that the lowest average annual value (for global alpha activity)) is recorded in 2018, while the highest annual average value is recorded in 2015 (see *Figure 4.6*).



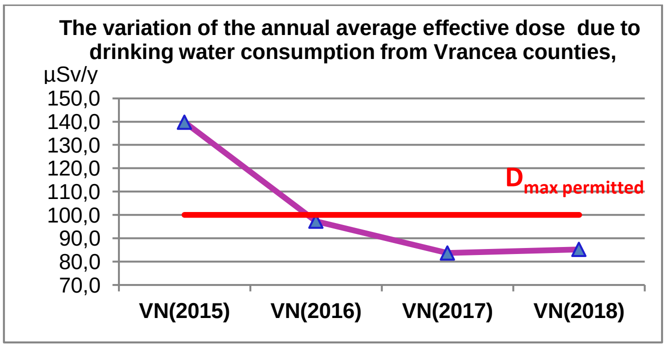


***Figure 4.6** – The variation of the radioactivity parameters (gross alpha, gros beta activity), and annual effective dose due to drinking water consumption from Vrancea county, during the period 2015-2018*

The annual average value of the gross beta activity registers a maximum in 2015 and a minimum in 2017. The dynamics regarding the use of water sources, their modernization or improvement probably induces a dynamic of the value ranges of the radioactivity parameters determined, Figure 4.6.

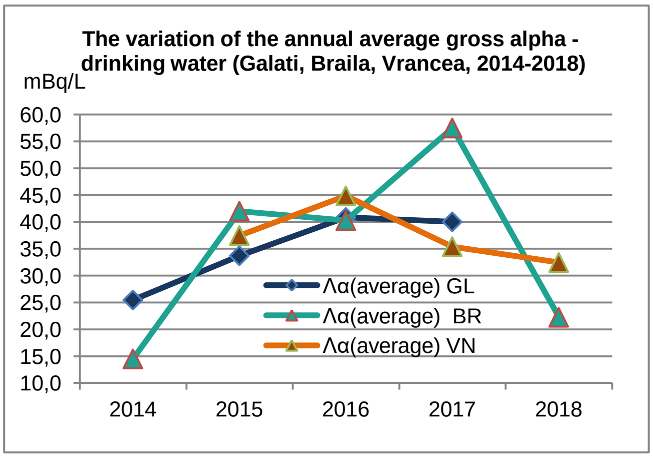
The value of the annual effective dose due to the consumption of drinking water evaluated for the period 2015-2018, in Vrancea county, varies in the range 21 ÷ 473 µSv year^{-1}, the average being 102 ± 19 µSv year^{-1}.

The risk due to the effective dose received by the population of Vrancea county through the consumption of drinking water, for the period 2015-2018, was evaluated at the value of 42.4x10^{-5}, slightly exceeded compared to 41.3x10^{-5}, the value of the risk calculated if the annual effective dose would reach the limit value of 100 µSv year^{-1}. In evaluating this risk coefficient was used: the average value of the annual effective dose determined in Vrancea county, for the period 2015-2018, of 102 µSv year^{-1}, the value of 75.70 years - representing the lifespan in Vrancea county, in the period 2015-2017, [http://statistici.insse.ro:8077/tempo-online - accessed on 12.12.2018] and the risk factor for cancer and genetic effects for the entire population (Sv^{-1}) of 5.7x10^{-2} Sv $^{-1}$ [ICRP, Publication 103].

Between 2014-2018, the gross alpha and gross beta activity determined in the drinking water collected from Brăila County, varies in the range 6 ÷ 75 mBq L^{-1}, and 25 ÷ 720 mBq /l, respectively. The annual effective dose due to the drinking water consumption, in Brăila County, evaluated for the period 2014-2018, based on formula (3.5) - (*Chapter 3, subchapter 3.1-thesis*), it led to an average value of 87 ± 17 µSv year^{-1}, within the value range of 18-428 µSv year^{-1}.

The risk due to the annual effective dose received by the population of Braila County through the consumption of drinking water, for the period 2014-2018, was evaluated at the value of 37.3x10^{-5}, placed below the value of 42.7x10^{-5}. For the average life span of Braila county, for the period 2014-2017, the value of 74.94 years was used, [http://statistici.insse.ro:8077/tempo-online - accessed on 12.12.2018]

During the period 2014-2018, in the region of Galați, Brăila and Vrancea counties, the highest average value of the global alpha activity (14,5÷57,5 mBq L^{-1}) was identified in the samples collected from Galați county, and then in the from Brăila county, followed by those from Vrancea county. The average values of the global beta activity (68.3÷223.5 mBq L^{-1}) depending on the sampling place, increase in the order: Galați, Brăila and Vrancea - variation shown in *Figure 4.7.*



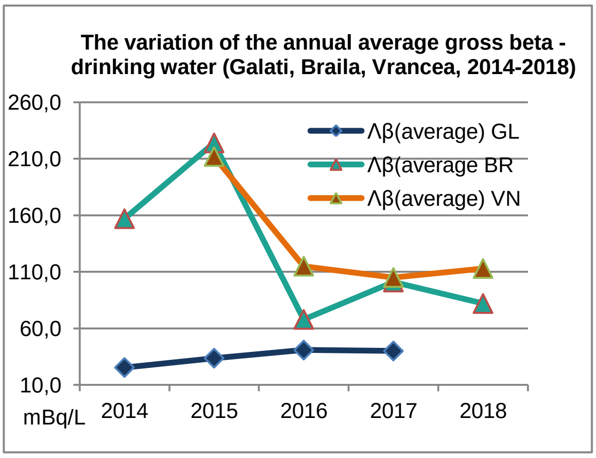


Figure 4.7 *- Variația activității alfa și beta global în apa potabilă în județele Galați, Brăila, Vrancea, în perioada 2014-2018*

It can be observed that in the world, both higher and lower values of global alpha and beta activity were determined compared to those determined in the present study.

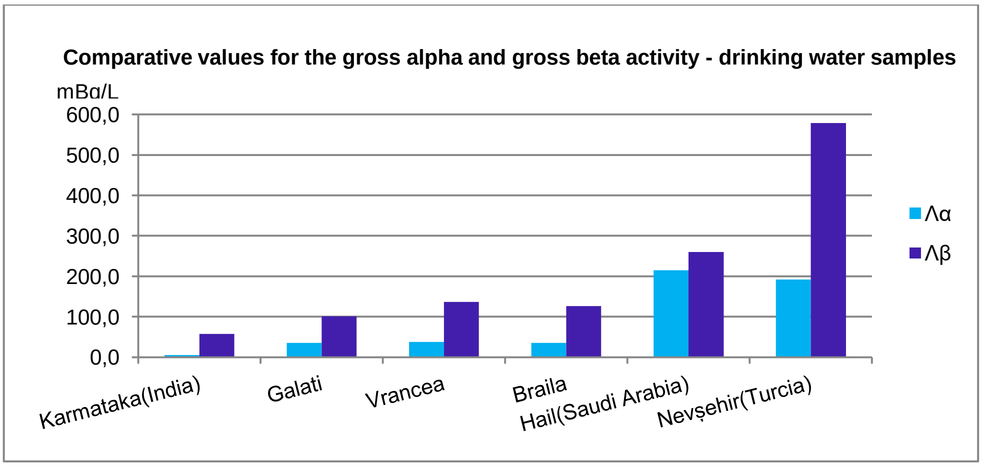


Figure 4.8 *– Comparative values for the gross alpha and beta activity – drinking water (the values from Galați, Brăila, Vrancea counties was determinatet during this study: 2014-2018)*

For all the analyzed samples collected in the period 2015-2018, from Galați, Brăila and Vrancea Counties, the distribution of the values of the gross alpha activities, respectively the gross beta, by value groups, is exemplified in *Figure 4.9.*

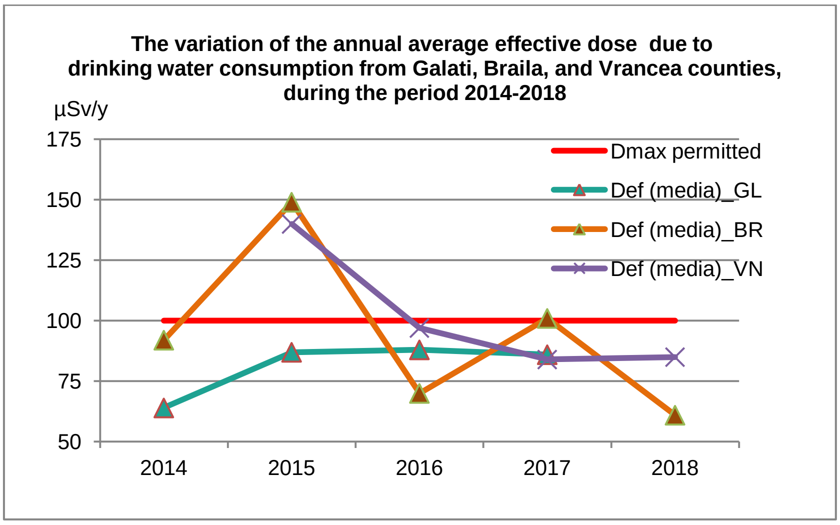


Figure 4.10 *– The variation of the annual effective dose due to drinking water consumption from Galați, Brăila and Vrancea counties, during the period 2014-2018*

It is noted that for 21% of the total drinking water samples, analyzed during the period 2014-2018, the values of the gross alpha activity ranged in the interval 0-10 mBq L^{-1}, while, for 61% of the samples, the gross beta activity has values in the range 0-100 mBq L^{-1}.

The annual effective dose due to the consumption of drinking water, evaluated in the period 2014-2018, in Galați, Brăila and Vrancea counties, varies in the range 18 ÷ 473 µSv $year^{-1}$. The variation in time of the determined annual averages of the annual effective dose is shown in *Figure 4.10.*

The annual effective dose due to the consumption of drinking water, evaluated on the basis of the gross alpha and gross beta activity, in the region of Galați, Brăila and Vrancea counties, between 2014 and 2018, varies as follows:

• for the population of Galați county, the annual effective dose due to drinking water consumption varies in the range 18÷222 µSv $year^{-1}$, with an average of 81 ± 14 µSv $year^{-1}$;

• for the population from Vrancea county, the annual effective dose varies in the range 21÷473 µSv $year^{-1}$, the average being 102 ± 19 µSv $year^{-1}$;

• for the population from Brăila county, the annual effective dose varies in the range 18÷428 µSv $year^{-1}$, with an average of 87 ± 17 µSv $year^{-1}$.

The detriment due to the radiation brought on the health of the population by the consumption of drinking water, evaluated for the period 2014-2018, for the population from the region of Galați, Brăila and Vrancea counties, varies as follows: $34.9x10^{-5}$ (compared to the maximum value of $42.8x10^{-5}$), $37.3x10^{-5}$ (compared to the maximum value of $42.7x10^{-5}$), respectively $44.8x10^{-5}$ (compared to the maximum value of $43.1x10^{-5}$).

In conclusion, the assessment of the annual effective dose due to the drinking water consumption in Galați, Brăila and Vrancea counties, between 2014 and 2018, based on gross alpha and beta activity, using the formula 3.5 (Chapter 3, sub-chapter 3.1.), leads to average values below the maximum allowed value (µSv $year^{-1}$) except that assessed for Braila county.

It is worth mentioning that this annual effective dose evaluation algorithm due to drinking water consumption, based on the global alpha and beta activity, leads to overestimation of the dose size, representing a maximum risk assessment. Therefore, for the evaluation of the annual effective dose the algorithm based on the concentrations determined by radionuclides was used (in the next paragraph, see also details in Chapter 3, Subchap 3.1).

The assessment of the annual effective reference dose based on the specific activities of ^{210}Po, ^{210}Pb, ^{nat}U, ^{226}Ra:

Based on the specific activities of ^{210}Po, ^{210}Pb, ^{nat}U, ^{226}Ra were evaluated:
- the annual effective dose due to drinking water consumption from Galați county, in 2015;
- the annual effective dose due to drinking water consumption from Vrancea county, during from 2015 to 2017;
- the annual effective dose due to drinking water consumption from Brăila county, during from 2016 to 2018;
- the annual effective dose due to mineral water consumption from Galați county, in 2015

In 2015 from Galați county, eleven samples of drinking water were analysed. From these the specific activities of ^{210}Po, ^{210}Pb, ^{nat}U, ^{226}Ra were determined.

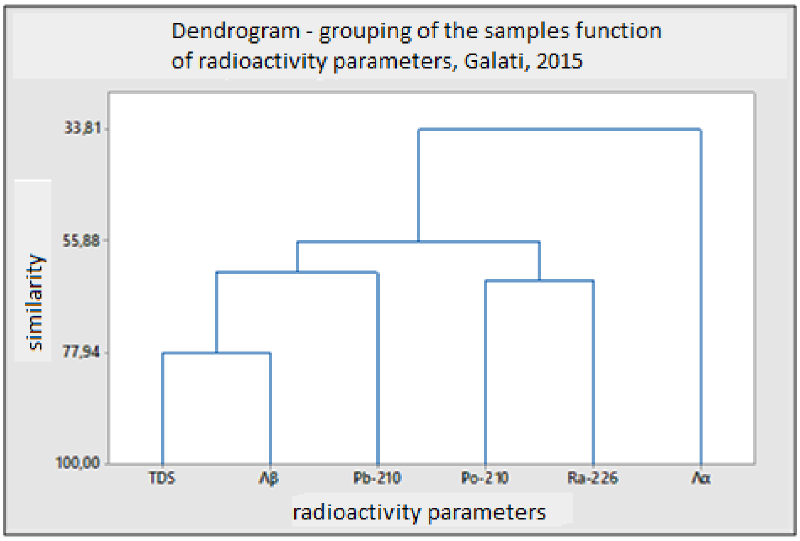


***Figure 4.11** – Grouping of the radioactivity parameters (gross alpha and beta activity, concentration of ^{210}Po, ^{210}Pb, ^{226}Ra) – drinking water samples collected from Galati, 2015*

The multivariate analysis determine the dendrograma from *Figure 4.11*. It is observed:

- ^{226}Ra and ^{210}Po with a similarity of 63.5%, are grouped in a cluster. ^{226}Ra, ^{210}Po are both alpha emitters and belong to the same natural radioactive series of ^{238}U;
- the total substance dissolved together with the gross beta activity with a similarity of 77.9% is grouped in a cluster, which connects with the parameter ^{210}Pb - with a similarity of 62.2%; ^{210}Pb is a beta emitter and belongs to the ^{238}U natural radioactive series, its determination is made by measuring the precipitate beta resulting from radiochemical separation.

The detriment due to the radiation brought on the health of the population through the consumption of drinking water, in Galati county, during 2015, led to the value of 5.3×10^{-5}, representing 12.8% of the damage due to the radiation if annual effective dose reached the maximum value of 100 µSv year^{-1}. For this evaluation were used: the average value of the annual effective dose corresponding to the drinking water consumption in Galați county, during 2015, of 12.3 µSv year^{-1}; the average life span of Galați county for the year 2015 of 75.21 years [http://statistici.insse.ro:8077/tempo-online - accessed on 12.12 2018] (*Table 4.7 – chapter 3, 3.2*) and the nominal risk coefficient adjusted to detriment to cancer and genetic effects (Sv^{-1}): $5{,}7x10^{-2}$ Sv^{-1}, [ICRP, Publication 103].

During the period 2015-2017 twenty-nine samples were analysed and the specific activities of ^{210}Po, ^{210}Pb, ^{nat}U, ^{226}Ra were determined. The multivariate analysis determine the dendrograma from *Figure 4.12*.

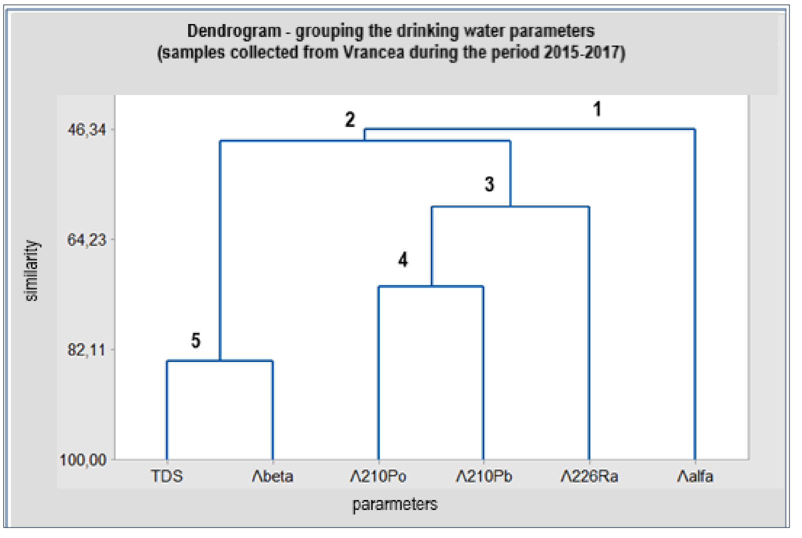


Figure 4.12 - *Dendrograma-grouping the drinking water parameters determined in samples collected from Vrancea, during the period 2015-2017*

The value indicators corresponding to the dendrogram in Figure 4.12, which indicates the number of clusters (clusters), the percentage of similarity between these groups, the correlation mode of the clusters, are shown in *Table 4.4*.

Table 4.4 - *Values indicators corresponding to the Dendrogram - parameters determined in drinking water, in Vrancea county, 2015-2017*

Level	No. cluster	Similarity	Distantce	Clusters connected		New Clusters	No. clusters Connected in new cluster
1	5	83,87	0,32	1	3	1	2
2	4	71,80	0,56	4	5	4	2
3	3	58,83	0,82	4	6	4	3
4	2	48,15	1,04	1	4	1	5
5	1	46,34	1,07	1	2	1	6

The grouping by clustering of the radioactivity parameters, determined in the drinking water from Vrancea county, between 2015 and 2018, indicates:

- belonging to the same group of variables: total dissolved substance and global beta activity (*cluster no. 5*) with a similarity of 83.3%, these in connected with the variable gross alpha activity with a much lower similarity of 46.3 % explained as follows: both gross beta activity and gross alpha activity are determined on the residue obtained after the primary processing of the sample. The final calculation of these parameters is influenced by the total dissolved substance value of the sample, more in the case of gross beta activity, in which case the beta particles emitted by the nucleus are characterized by a continuous energy spectrum, and less in the case of the gross alpha activity, in the case which is very small during alpha radiation.

- belonging to the same group of variables Λ_{210Po} and Λ_{210Pb}, with a fairly high percentage of similarity, of 71.8%, these connected in turn with the variable Λ_{226Ra}, this fact is explained as follows: the radionuclide ^{210}Po, is produced by the disintegration of the radionuclide ^{210}Pb and of course, the ^{210}Pb content in the sample is highly dependent on the ^{210}Po content. In fact, some radiochemical methods for determining ^{210}Pb in water are precisely based on this parent radionuclide (^{210}Pb) - daughter radionuclide (^{210}Po) bond. The aforementioned radionuclides together with ^{226}Ra, with which a group is observed are components of the same natural radioactive series - the Uranium -238 series.

In the drinking water sampled in the period 2015-2018, from Galați, Brăila and Vrancea counties, the concentration of 210Po ranges from 0.5 to 12.5 mBq L^{-1} the average being 2.7 ± 0.6 mBq L^{-1} and the median having a value of 2.0 ± 0.4 mBq L^{-1}. while for the 210Pb concentration an average of 4.7 ± 0.7 mBq L^{-1} is determined in the range of 0.6÷19.5 mBq L^{-1}, characterized by a median of 3.4 ± 0.5 mBq L^{-1}. The distribution of the values of concentrations of ^{210}Po and ^{210}Pb from drinking water, in Galați Brăila, Vrancea counties, between 2015-2018, is illustrated in *Figure 4.13.*

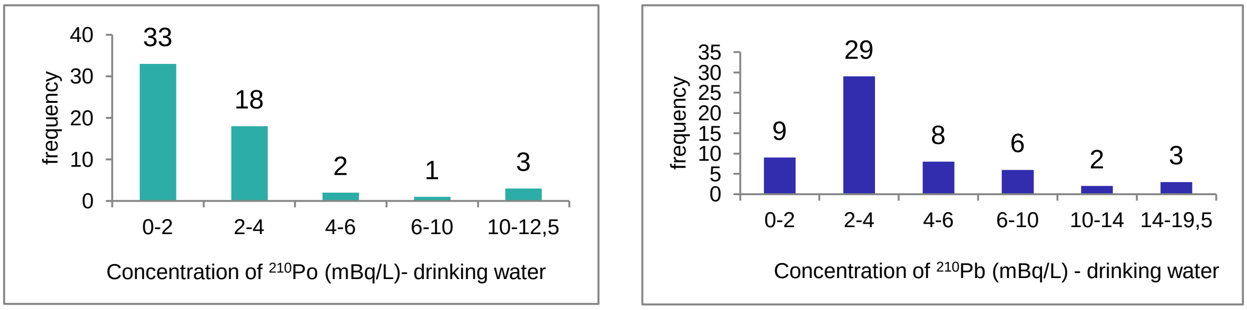
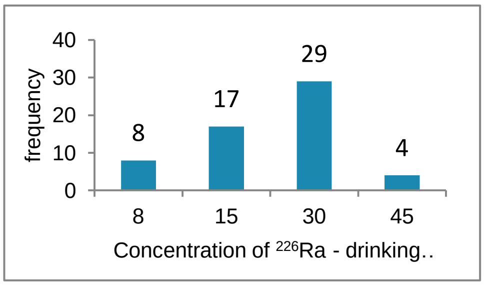


Figure 4.13 *– Distribution valueas of the radioactivity parapeters (concentration of ^{210}Po and ^{210}Pbfrom drinking water samples, collected from Galați, Brăila, and Vrancea, during the period 2015-2018*

For drinking water from Karnataka (India), values for ^{210}Po concentration in the range of 1.89-4.18 mBq L^{-1} are mentioned, with an average of 3.22 mBq L^{-1}, [Kavitha E., 2017]. Much wider range of values (0-114.2 mBq L^{-1}) - for ^{210}Po concentration, it is determined for drinking water from Western Australia [Walsh, 2014], while for ^{210}Pb concentration, the same source indicates similar values of the present study (LLD -13.4 mBq L^{-1}, where LLD – low limit detection). Much lower values for ^{210}Po, and ^{210}Pb, in the ranges: 0.25-0.7 (0.6 ± 0.1) mBq L^{-1}, and 0.7-2.7 (1.9 ± 1.5) mBq L^{-1}, respectively are determined [Rožmarić M., 2012] in mineral waters from Croatia. These much lower values could be due either to the lower composition in radionuclides, or to the time elapsed between bottling and radiochemical determinations, given the relatively short half-lives for ^{210}Po ($T_{1/2}$ = 138 days) and ^{210}Pb ($T_{1/2}$ = 22.3 years).

In the drinking water of Germany the concentration of ^{210}Po is in the range 0.2-180 mBq L^{-1} (with a median of 1.4 mBq L^{-1}), [Beyermann, 2019], with a maximum, much higher than the maximum of the present study. .



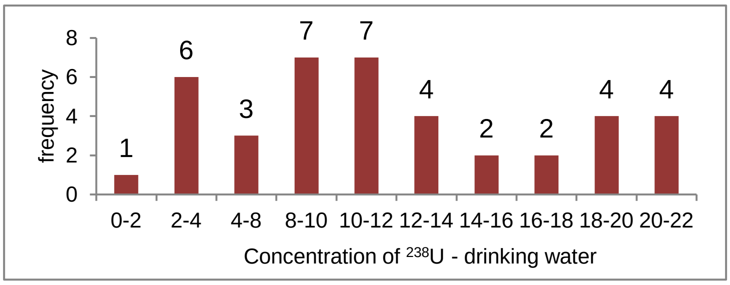


***Figure 4.14** - Distribuția valorilor concentrațiilor de ^{226}Ra și ^{238}U (mBq/l) din apă potabilă, în județele Galați Brăila, Vrancea, în perioada 2015-2018, pe grupe de valori*

In Galați, Brăila and Vrancea counties, in the period 2015-2018, the water samples have a content of ^{226}Ra between 8 and 45 mBq L^{-1}, the average being 18.5 ± 5.6 mBq L^{-1}. For drinking water in Kumasi (Ghana) an average of 22.41 mBq L^{-1} is determined, [Darko G., 2015]. Neighboring values are reported on both sides of the domain determined in this study for the concentration of ^{226}Ra, namely: 6.43-12.59 mBq L^{-1} in drinking water from Ondo (Nigeria) and 2.08-78.36 mBq L^{-1} in drinking water from Ekiti (Nigeria), [Ayodele AE, 2017]. Very high values of ^{226}Ra in drinking water are determined in Ramsar (Iran): 16-524 mBq L^{-1}, [Farthabadi S., 2019], Ramsar being among the areas with the highest natural radioactivity on Earth, [Farthabadi S., 2017]. The value distribution of the concentrations of ^{226}Ra and ^{238}U from drinking water, in Galați, Brăila and Vrancea counties, in the period 2015-2018, is shown in *Figure 4.14.*

The concentration of ^{238}U determined in the drinking water samples, from Galați, Brăila and Vrancea counties, during the period 2015-2018, has values in the range 1.0-22.0 mBq L^{-1}, the average being 11.9 ± 1, 3 mBq L^{-1}. Slightly lower values, from 0 to 14.3 mBq L^{-1} with an average of 2.3 mBq L^{-1} are determined in drinking water in Western Australia, [Walsh, 2014]. Much higher maximums are recorded for mineral waters in Italy in the range 0.206-103 mBq L^{-1} (average 21.4 mBq L^{-1} [Jia G., 2009],

The average value of the annual effective dose, received by the adult population through drinking water consumption, evaluated in the region of Galați, Brăila and Vrancea counties, in the period 2015-2018, is 10.69 ± 1.3 μSv year^{-1}. It is composed of: the effective dose due to ingestion ^{210}Po, 4.1 ± 0.5 μSv year^{-1}, the effective dose due to the ingestion ^{210}Pb, 2.4 ± 0.4 μSv year^{-1}, the effective dose due to the ingestion ^{226}Ra, of 3,8 ± 0.4 μSv year^{-1} and the effective dose due to ingestion of ^{238}U, 0.39 ± 0.04 μSv year^{-1}, as illustrated in *Figure 4.17.*

The largest contribution at the annual effective dose brought to the population from Galați, Brăila and Vrancea counties by drinking water consumption is brought by the ingestion of radionuclide ^{210}Po, followed by that of ^{226}Ra, ^{210}Pb and then ^{238}U. It is noted that the most important radionuclides, in terms of contribution to the annual effective dose, are ^{210}Po, ^{226}Ra, ^{210}Pb. The smallest contribution to the annual effective dose due to the consumption of drinking water is the intake of the ^{238}U isotope, with a percentage of only 4%.

Some authors consider that, in addition to the contribution of the aforementioned radionuclides and the contribution of the primordial radionuclide ^{40}K [Jia G., 2009] should be considered. This would contravene the calculation European rules [European Directive 51/2013] and its transposition into national law [Law 301/2015], regarding the establishment of the requirements of protection of the health of the population regarding the radioactive substances in the drinking water.

Contribution of radionuclides to the annual effective dose due to drinking and mineral water consumption is ilustrated in *Figure 4.17* and *Figure 4.17*.

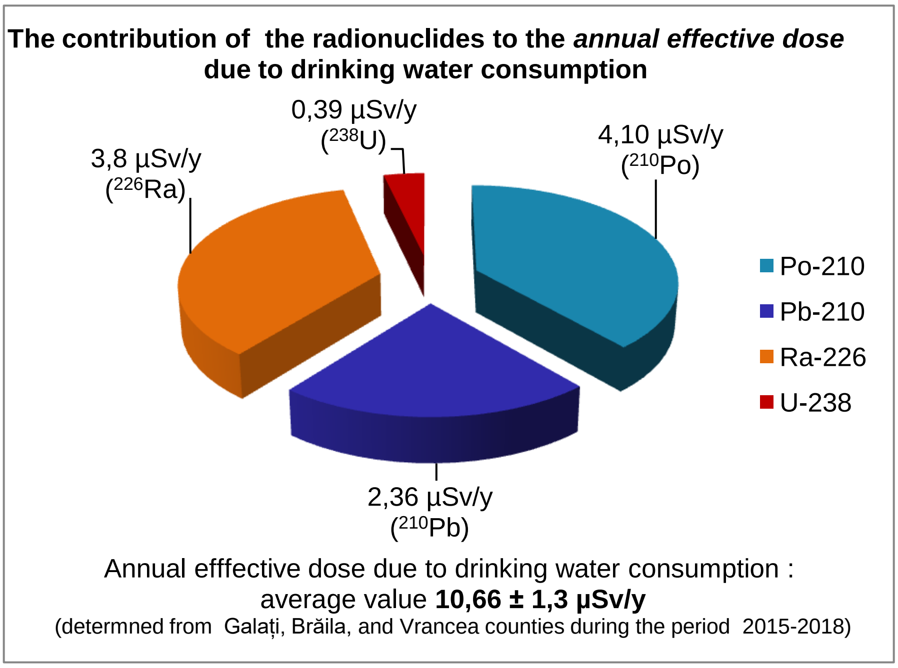


Figure 4.17 *– The contribution of the radionuclides ^{210}Po, ^{226}Ra, ^{210}Pb, ^{238}U to the annual effective dose due to drinking water consumption assessing from Galați, Brăila, and Vrancea counties during the period 2015-2018*

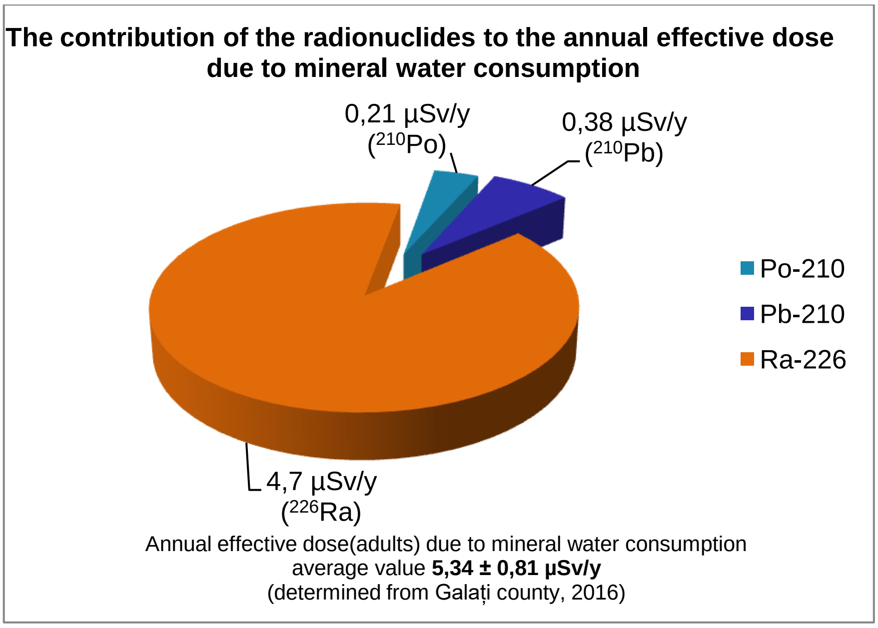


Figure 4.18 *- The contribution of the radionuclides ^{210}Po, ^{226}Ra, ^{210}Pb, to the annual effective dose due to mineral water consumption (samples collected from Galați county, 2016*

The average of the annual effective dose due to the ingestion of radionuclides ^{226}Ra, ^{210}Po and ^{210}Pb from mineral water consumption is 5.34 ± 0.81 µSv year^{-1}, in the case of this study, representing half of the dose brought to the human body by drinking water consumption.

For the evaluation of the annual effective dose due to the consumption of mineral water, the gross alpha, gross beta activity, and the concentration of ^{210}Po, ^{210}Pb and ^{226}Ra were determined. The values of the radioactivity parameters determined in the mineral water are compared with the values determined in the countries, *Table 4.5.*

Table 4.5 *- Concentration of radionuclides in mineral water in different countries (mBq L^{-1}): Gross alpha activity (Λ_α), gross beta activity (Λ_β), concentration of ^{226}Ra (Λ_{226Ra}), concentration of ^{210}Po (Λ_{210Po}), concentration of ^{210}Pb (Λ_{210Pb}), min-max, (average)*

Country	Λ_α	Λ_β	Λ_{226Ra}	Λ_{210Po}	Λ_{210Pb}	References
	min-max (average) (mBq L^{-1})					
Bucovina, Romania	0.40–45.40 (12.13)	1.51–47.45 (11.34)				[Călin M. R., 2016]
Serbia		–	1–13	–	41–173	[Janković M. M., 2012]
Croatia			0,67-38,1 (6,8)	0,51-3 (1,3)	0,8-7,6 (3,8)	[Rožmarić M., 2012]
Belgia, France Italy, Polonia	(329.3)		1.5-632	1.5-10.8		[Jobbágy V., 2013]
Ungaria	35-1749 (189)	33-2015 (209)				[Jobbagy V., 2011]
Turkey			<0.56-165			[Erden P.E., 2014]
Turkey	(125)	(170)	(129)			[Kobya Y., 2011]
Slovenia			0.14-32 (10.57)	0.24-2.1 (0.86)	0.6-13.2 (0.86)	[Benedik L., 2012]
Bucovina, Romania	1.03-5.50	15.9-31.4	110-450	-	-	[Călin M. R., 2019]
România	9-81 (17,9)	85-659 (198)	5-70 (33)	0.02-0.97 (0.34)	0.151-2.85 (1.10)	present study

The values of the natural radionuclides concentration are different due to of the geological structure of the areas of origin of the water sources

The contribution of radionuclide ^{226}Ra to *the annual effective dose* due to the consumption of mineral water is similar in order of magnitude as in the case of drinking water consumption, but first, as a contribution to the total effective dose.

Concentrations of ^{210}Po and ^{210}Pb respectively are lower in mineral water than that determined in drinking water, the reason for this is the time elapsed between bottling and determinations, knowing that this radionuclide has a relatively short half-life (T = 138.38 days).

The detriment due to ionizing radiation, brought to the health of the population from Galați county, by the consumption of mineral water in 2016, is evaluated at 2.3×10^{-5}, which represents 5.1% of the value of the radiation damage, if the annual effective dose due to drinking water consumption reaches the maximum permissible value of 100 µSv year^{-1}.

Based on the above results it is concluded that both drinking water, from Galați, Brăila and Vrancea counties, as well as the mineral water marketed in Galați is safe for population consumption, from the point of view of the content in natural radionuclides and the contribution to the annual effective dose.

3.2. Assessment of the annual effective dose due to foodstuffs consumption

The purpose of this research field was three-fold: (1) to determine the gross alpha activity, the gross beta activity and the concentrations of ^{210}Po, ^{210}Pb, ^{238}U, ^{232}Th, ^{226}Ra, and ^{40}K in foodstuffs samples collected from the three counties of Romania (Galati, Braila and Vrancea); (2) to evaluate the annual effective dose due to the ingestion of radionuclides through the foodstuffs consumption; and (3) to estimate the lifetime risk correspondent to the foodstuffs consumption.

The assessment to the annual effective dose due to bread consumption

To the best of our knowledge, the information regarding the natural radionuclides in bread is very scarce in literature and no published data on their concentrations is currently available for Romania, most authors reporting the content of radionuclides in flour, biscuits, and flour products [Abojassim A. A., 2014],[Abojassim A. A., 2015a], [Abojassim A. A., 2015b], [Turtiainen T., 2011], [Meli M. A., 2014]. In the scientific literature, [Meli M..A, 2014] were found for the concentration of ^{210}Po values in the range 0.020÷0.114 Bq kg^{-1} for flour samples collected from Italy, similar in the beginning part of the concentration range (^{210}Po) determined in the present study (0.014÷0.031 Bq kg^{-1}), but with a higher average namely 0.061 ± 0.026 Bq kg^{-1} compared to the present study.

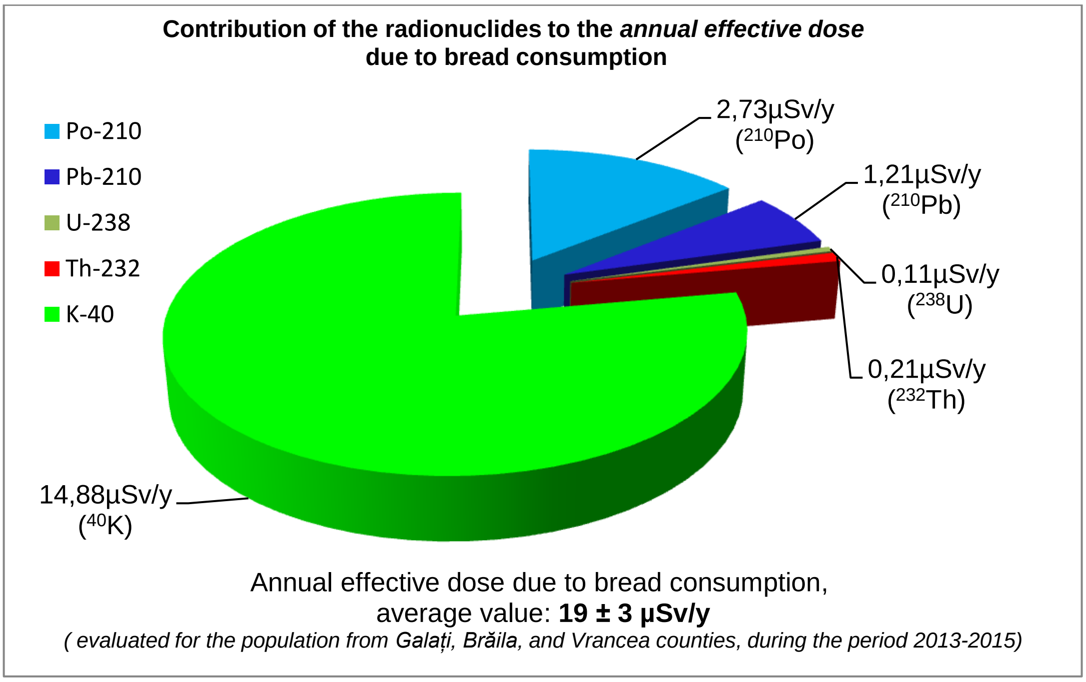


***Figure 4.19** – The contribution of the radionuclides ^{210}Po, ^{210}Pb, ^{238}U, ^{232}Th, and ^{40}K. to the annual effective dose due to bread consumption evaluated for the population from Galați, Brăila and Vrancea counties, during the period 2013-2015*

In the UNSCEAR report [Unscear, 2000] it was declared for wheat samples, the value range for ^{210}Po concentration of: 0.020÷0.360 Bq kg^{-1}. In white flour, colectted from Finland, [Turtiainen T., 2011], the concentration of ^{210}Pb is in a higher value range (0.058÷0.300 Bq kg^{-1}) compared to the area of the present study (0.006÷0.080 Bq kg^{-1}) and compared to the value field, for wheat, published in the UNSCEAR report [Unscear, 2000].

For the concentration of ^{238}U, in the present study, values in the range 0.009÷0.040 Bq kg^{-1} were determined. For wheat, the UNSCEAR report [Unscear, 2000] shows slightly higher ^{238}U values. For biscuits, in Iraq, ^{238}U values were reported in the range: 0.009÷0.040 Bq kg^{-1}, [Abojassim A. A., 2014]. It should be mentioned that *Abojassim et al.*, determined ^{238}U by gamma spectrometry with NaI(Tl) detector , in which case the sensitivity of the method is of the order Bq, compared to the sensitivity of the radiochemical methods which is of the order mBq. For the concentration of ^{232}Th, the value range determined in this study of 0.001÷0.021 Bq kg^{-1} is similar to that in the UNSCEAR report: 0.0016÷0.033 Bq kg^{-1}. A much higher value domain, explained by using the principle of gamma-spectrometric measurement (with NaI(Tl) detector), for the same radionuclide, is obtained by *Ashahri et. al.:* 5.6÷26 Bq kg^{-1} [Alshahri F., 2016] . *Tufail et. al.* obtain values, for ^{232}Th, in the range 1.0÷1.5 Bq kg^{-1} [Tufail M., 2010] using a gamma-spectrometer with a HpGe, high pure germanium detector. *Abojassim et al.* [Abojassim A. A., 2014], determined ^{232}Th by gamma spectrometry with NaI(Tl) detector, obtaining in flour samples values in the range: 0,126÷4,298 Bq kg^{-1}. For ^{40}K, in this study, values in the range 10.4÷31,4 Bq kg^{-1} were obtained. In flour samples, in Spain, values are obtained, for ^{40}K, in the range: 67÷122,7 Bq kg^{-1}, [Ballesteros L., 2015a]

In Poland, for wheat samples, ^{40}K ranged from 127.9 to 145.1 Bq kg^{-1} [Solecki J., 2011b], while for the south of India, the average of ^{40}K reported is 482.7 Bq kg^{-1}, [Shanthi G., 2010]. For wheat flour samples collected from Iraq, for ^{40}K values is reported the interval 41.84÷264.72 Bq kg^{-1}, [Abojassim A. A., 2014]. In white bread samples, coectted from Saudi Arabia, values of ^{40}K are determined, in the range 203÷297 Bq kg^{-1}, [Alshahri F., 2016]. In wheat flour samples from Pakistan, ^{40}K values are reported in the range 95.7÷146.9 Bq kg^{-1}, [Tufail M., 2010].

The evaluation of the annual effective dose due to the ingestion of the radionuclides ^{210}Po, ^{210}Pb, ^{238}U, ^{232}Th, and ^{40}K, through the consumption of bread, for the region of Galaţi, Brăila, Vrancea counties led to the average value of 19 ± 3 µSv year^{-1} and it was noted that, the largest contribution to this belongs to the ^{40}K isotope, *Figure 4.18.*

The average value of the annual effective dose due to the consumption of bread, evaluated in this study is much lower than the value of 110 µSv year^{-1}, representing the annual effective dose due to the ingestion of radionuclides from the uranium and thorium series, derived from the adult food and water consumption, evaluated by UNSCEAR, [Unscear, 2000].

Risk assessment, as a detriment due to the ionizing radiation brought to the population, through the consumption of bread, taking into account the average life span, during the period 2013-2015 in Galati, Braila, Vrancea counties (73.3 years) [http: //statistici.insse .ro / shop /, accessed on 12.12.2016], leads to the value of 8.1x10^{-5}, well below the value of 10.3x10^{-3}, representing the detriment due to all the natural sources evaluated for the region of Galați, Brăila and Vrancea counties, between 2013-2015.

The exposure assessment due to milk consumption

Pasteurized milk, one of the commonly foods found in human nutrition, brings into the human body the ^{40}K radionuclide, primordial radionuclide as well as natural radionuclides from the three natural radioactive series.

Table 4.6 *– Pearson correlation – radioactivity parameters (gross alpha activity (Λ_α), gross beta activity (Λ_β), concentration of ^{226}Ra (Λ_{226Ra}), concentration of ^{210}Po (Λ_{210Po}), concentration of ^{210}Pb (Λ_{210Pb}), concentration of ^{232}Th (Λ_{232Th}), concentration of ^{238}U (Λ_{238U}), concentration of ^{40}K (Λ_{40K})) determined in pasteurized milk*

Radioactivity parameters	Λ_α	Λ_β	Λ_{210Po}	$\Lambda_{210\backslash Pb}$	Λ_{238U}	Λ_{232Th}	Λ_{40K}
Λ_α	1						
Λ_β	-0,14	1					
Λ_{210Po}	-0,50	-0,26	1				
$\Lambda_{210\backslash Pb}$	-0,22	0,16	0,40	1			
Λ_{238U}	0,44	0,45	**-0,57**	0,46	1		
Λ_{232Th}	**0,73**	-0,15	**-0,64**	-0,55	0,30	1	
Λ_{40K}	0,28	0,08	-0,40	0,43	**0,80**	0,38	1

$p<0{,}05$

The correlation between the concentrations of radionuclides ^{210}Po ^{210}Pb, ^{232}Th, ^{40}K, ^{238}U, determined in pasteurized milk in this study, and shown in Table 4.6, indicated the following:

• a high positive correlation between ^{40}K, ^{238}U, explained by the fact that both primordial radionuclides, with very large half-lives ($T_{1/2-4-40K}$=1.28 x 10^9 years, $T_{1/2-4-238U}$= 4,5 x 10^9 years) existing in all the natural components of the environment that are transferred from the animal feed into their products;

• a high negative correlation between radionuclides ^{238}U, ^{210}Po, is explained as follows: radionuclide ^{210}Po is produced by disintegration of the natural radioactive family of uranium-238; therefore within a matrix of fresh product, as the radionuclide ^{238}U disintegrates, the concentration in the generated radionuclides, including the radionuclide ^{210}Po, increases;

• a high positive correlation between alpha activity and the concentration of ^{232}Th, due to the fact that ^{232}Th is the alpha emitter;

• A reasonable positive correlation is observed between the radionuclide ^{238}U and the global alpha and beta activity, explained by the fact that the parent radionuclide ^{238}U generates by decay both alpha and radionuclide beta emitters.

In Rostov (Russia) in milk samples, concentrations of 0.008 Bq kg^{-1} and 0.020 Bq kg^{-1} were found for ^{210}Po and ^{210}Pb, respectively [Ladinskaya L., 2013], compared to the average values determined in this study, namely : 0.041 ± 0.012 Bq L^{-1} for ^{210}Po and 0.016 ± 0.004 Bq L^{-1}. The specific activity of ^{40}K is evaluated at 43 Bq kg^{-1} in Korea [Chae J.S., 2016], at 34 Bq kg^{-1} in India [Shanti G., 2010], at 56.8 ± 13.3 Bq kg^{-1} in the Netherlands [Brandhoff P.N., 2016], in the range 17.7÷72.37 Bq L^{-1} in Spain [Ballesteros L., 2015], compared to the average value of 45.48 ± 6.82 Bq L^{-1}, determined in this study.

In Ramsar, Iran, an area with increased natural radioactivity, [Sohrabi M., 2013], [Fathabadi N., 2017], the annual effective dose due to the ingestion of radionuclide ^{226}Ra was evaluated, at the maximum value of 1.52 µSv $year^{-1}$, the average value being of 0.93 µSv $year^{-1}$, [Fathabadi N., 2019]. In Korea, a contribution of 1182 Bq year^{-1} was evaluated due to the ingestion of ^{40}K radionuclide, which generates an effective annual dose of 7.3 µSv year^{-1}, [Chae J.S., 2016].

The evaluation of the annual effective dose due to the ingestion of radionuclides ^{210}Po, ^{210}Pb, ^{232}Th, ^{40}K, ^{238}U by the pasteurized milk consumption evaluated for adults, in the region of Galați, Brăila and Vrancea counties, between 2014 and 2016, led to an average value of 24.99 ± 3.25 µSv year^{-1}, the contribution of the radionuclides to this being exemplified in *Figure 4.21.*

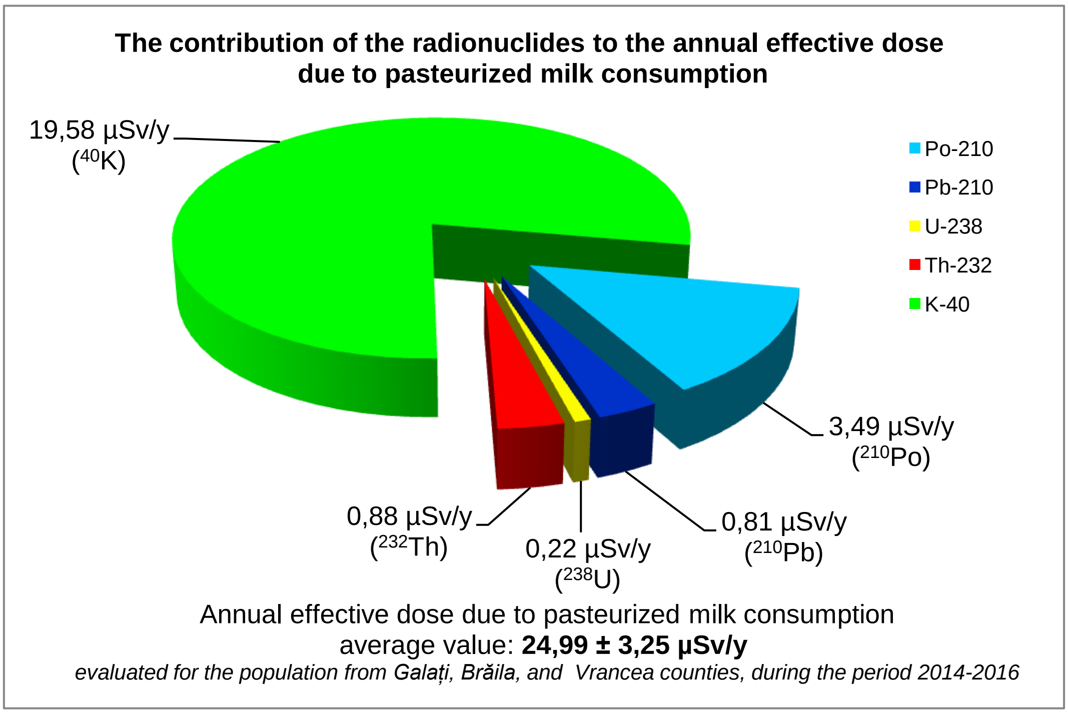


Figure 4.21 *– The contribution of radionuclides ^{210}Po, ^{210}Pb, ^{238}U, ^{232}Th, ^{40}K, from pasteurized milk to the annual effective dose, evaluated for the population from Galați, Brăila,and Vrancea counties, during the period 2014-2016*

The highest contribution to the annual effective dose due to the consumption of pasteurized milk belongs to the ^{40}K radionuclide, with a percentage of 78.35%, while the lowest contribution is due to the radionuclide ^{238}U, with a percentage of 0.88%. The radionuclides ^{232}Th, ^{210}Pb, ^{210}Po contribute to the annual effective dose with a percentage of 3.52%, 3.24%, respectively 13.96%.

In the case of pasteurized milk samples, the concentration of ^{210}Po is higher than the concentration of ^{210}Pb, which is the opposite of the case of powdered milk samples intended for children in the 0-12 month age category. This can be explained on the basis of the following reasoning: ^{210}Pb disintegrates into ^{210}Bi and then at ^{210}Po, the relatively short half-life of the mentioned descendants ($T_{1/2\text{-}Bi\text{-}210}$=5 days, $T_{1/2\text{-}Po\text{-}210}$=138 days) is not enough to establish secular balance, in fresh samples with short processing time. The disintegration scheme of ^{210}Po to stable ^{216}Pb is described below [Hou X., 2008]:

$$^{210}\text{Pb} \xrightarrow[T_{1/2}=\,22,3\text{ y}]{\beta} {}^{210}\text{Bi} \xrightarrow[T_{1/2}=5\text{ d}]{\beta} {}^{210}\text{Po} \xrightarrow[T_{1/2}=138,4\text{ d}]{\alpha} {}^{216}\text{Pb (stable)}$$

In the case of powdered milk samples in addition to the longer processing time, the longer shelf life, which contributes to the increase of the disintegration time of ^{210}Pb to ^{210}Po, also occurs the processing temperature, it is known that at temperatures above 80°C, ^{210}Po is volatilized, so that the drying and processing of the powdered milk contributes to the decrease of the concentration of ^{210}Po. Moreover, it is very possible that in the additives to the powder additive there was not enough time to achieve the secular equilibrium ^{210}Po/^{210}Pb, which contributes to the increase of the concentration of ^{210}Pb from that of the radionuclide ^{210}Po.

The assessment of the risk as detriment due to the ionizing radiation brought to the population, through the consumption of pasteurized milk, evaluated in the region of Galați, Brăila and Vrancea counties, in the period 2014-2016, led to values in the interval 8.01x10^{-5}-13,32x10^{-5}, with an average of 10.71x10^{-5}. In this evaluation, the average human lifespan of Galați, Brăila and Vrancea counties, for the period 2014-2016, was taken into account (http://statistici.insse.ro/shop/, accessed on 18.01.2017) and the coefficient nominal risk adjusted for cancer and genetic effects recommended by Publication 103-ICRP, amounting to 5.7 x 10^{-2} Sv^{-1}.

Table 4.7 *- Average life span for the population from Galați, Brăila and Vrancea counties, between 2014-2017*

County	2013	2014	2015	2016	2017
	Average life span				
Galați	74,65	74,89	75,21	75,53	75,18
Brăila	74,44	74,65	74,62	75,29	75,22
Vrancea	75,55	75,49	75,54	75,84	75,73

http://statistici.insse.ro/shop/

The value of the risk thus calculated is well below the value of 10.29x10^{-3}, representing the detriment due to all natural sources of 2.4 mSv year^{-1} [UNSCEAR, 2000], evaluated for the region of Galați, Brăila and Vrancea counties, in 2015 -2016.

The exposure assessment due to meat consumption

During 2014–2016, various types of meat (fish, pork, chicken) were collected from various markets of Galati, Braila and Vrancea. Twenty-seven meat samples, were investigated in order to assess the effective dose from the natural radionuclides ^{210}Po, ^{210}Pb, ^{238}U, ^{232}Th, ^{226}Ra, and ^{40}K.

The literature is very poor regarding the gross alpha and gross beta activity from meat samples. Zorer and Öter (2015) reported for the gross alpha activity the values: 0.625 ± 0.379 Bq g^{-1} (sample ashed at 600˚C) for fresh fish and undetectable values for salted fish, and for gross beta activity the value 2.863±1.096 Bq g^{-1} (sample ashed at 600˚C) for fish (chalcalburnus tarichi) [Zorer Ö. S. 2015].

In this study the highest concentration of ^{210}Po was determined in fish, the average value being 52 ± 7 mBq kg^{-1}, which is lower than the values reported in literature. Šrok and Smodiš (2011) obtained the concentration values for ^{210}Po in freshwater fish in the range 56 - 180 mBq kg^{-1}; the concentration of ^{210}Po for freshwater fish species is lower than marine fish species [Štrok M., 2011]. Higher values of the ^{210}Po concentration were reported by Khan and

Wesley (2015) for tuna fish, ranging from 40.09 ± 5.2 Bq kg^{-1} to 92.5 ± 7.9 Bq kg^{-1} (average 65.55 Bq kg^{-1}) [Khan M. F., 2016]. Aoun et al. (2015) reported for ^{210}Po concentration the following values: 1.9 ± 0.2 Bq kg^{-1} and 46.9 ± 3.5 Bq kg^{-1} for fish of the species *Erytrinus*, and *P. Rivulatus*, respectively [Aoun M., 2015].

With the aid of specific activity of this nuclide in water, and the concentration ratio (bioaccumulation factor) of 36 L kg^{-1} f.w. for freshwater fish tissues (muscles) published by IAEA (2010) [IAEA, 2010], the prediction of water-biota radionuclide transfer has been assessed in this work; the reported specific activity of ^{210}Po for drinking water in Galati region in 2015, having as source the Danube river - the same source for local fish - was 2.0 ± 0.4 mBq L^{-1} [Pintilie V., 2016b]. Using these values, the prediction of the ^{210}Po concentration in fish is 72 mBq kg^{-1}, which is comparable with the values of the present study (52 ± 7 mBq/kg)..

The average value of the ^{210}Po concentration in pork samples obtained in this study was 24 ± 3 mBq kg^{-1} f.w. (fresh weight), which is similar to that reported by [Meli M. A., 2013] for pork samples collected in Italy (20 ± 10 mBq kg^{-1} f.w.). In this study, the concentration of ^{210}Pb ranged from 7 ± 1 to 21 ± 3 mBq kg^{-1} for the fish samples, from 8 ± 1 to 28 ± 4 mBq kg^{-1} for pork samples, and from 9 ± 1 to 21 ± 3 mBq kg^{-1} for chicken samples. In [Aoun M., 2015] there were reported ^{210}Pb concentrations for fish collected from Byblos Lebanese coastal zone ranging from BDL (below the detection limit) to 18.2 Bq kg^{-1}.

The concentrations of ^{238}U ranged from 9 ± 1 to 37 ± 3 mBq kg^{-1} for the fish samples, from 10 ± 1 to 58 ± 5 mBq kg^{-1} for the pork samples, and from 12 ± 1 to 57 ± 5 mBq kg^{-1} for the chicken samples. The concentrations of ^{232}Th ranged from 5 ± 0.5 to 12 ± 1 mBq kg^{-1} for the fish samples, from 6 ± 1 to 19 ± 2 mBq kg^{-1} for the pork samples, and from 9 ±1 to 17 ± 2 mBq kg^{-1} for chicken samples. Pietrzak-Flis et al. (1997) [Pietrzak-Flis Z., 1997] found lower values for the concentrations of ^{238}U and ^{232}Th in the pork samples collected from Walbrzych (Poland): 1.68 ± 0.46 mBq kg^{-1} and 0.52 ± 0.19 mBq kg^{-1}, respectively.

The concentrations of ^{40}K ranged from 51 ± 9 to 126 ± 21 Bq kg^{-1} for the fish samples, from 41 ± 7 to 56 ± 10 Bq kg^{-1} for the pork samples, and from 18 ± 3 to 30 ± 5 Bq kg^{-1} for the chicken samples. Ballesteros et al. (2015) [Ballesteros L., 2015] found the concentration of ^{40}K ranging from 58 to 158.9 Bq kg^{-1} for fish samples. Shanthi et al. (2010) [Shanthi G., 2010] reported for fish samples 88.91 ± 6.7 Bq kg^{-1}, f.w. the specific activity of ^{40}K. Chae et al. (2016) [Chae J. S., 2016] reported the average value of the ^{40}K concentration of 85 ± 34 Bq kg^{-1} (ranging from 48 Bq kg^{-1} to 178 Bq kg^{-1}) for pork, and the average value of 69 ± 16 Bq kg^{-1} (ranging from 44 Bq kg^{-1} to 94 Bq kg^{-1}) for chicken.

The concentrations of ^{226}Ra obtained in this work ranged from 0.7 ± 0.1 to 5.7 ± 0.7 Bq kg^{-1} for the fish samples, from 3.1 ± 0.4 to 4.0 ± 0.5 Bq kg^{-1} for the pork samples, and from 2.4 ± 0.3 to 4.6 ± 0.6 Bq kg^{-1} for chicken samples. Shanthi et al. (2010) [Shanthi G., 2010] reported for fish samples a concentration of ^{226}Ra of 0.05 ± 0.01 Bq kg^{-1} (f.w.). Pietrzak-Flis et al. (1997) [Pietrzak-Flis Z., 1997] found an activity of 11.6 ± 1.36 mBq kg^{-1} f.w. for pork samples (Walbrzych region-Poland). It is worth mention that the concentration of ^{226}Ra in water in that area was 1.70 ± 0.15 mBq L^{-1} [Pietrzak-Flis Z., 1997], which is lower than the concentration of ^{226}Ra in water in the area of the present study (30 ± 9 mBq L^{-1}) reported in [Pintilie V., 2016b].

The concentrations of radionuclides in meat samples ranged in different parts of the world depending on the used feed and livestock farming [UNSCEAR, 2000].

The annual effective dose due to ingestion of radionuclides ^{210}Po, ^{210}Pb, ^{238}U, ^{232}Th, ^{226}Ra, and ^{40}K through meat consumption, ranged from 3.4 ± 0.5 to 13.2 ± 1.9 µSv y^{-1} for fish samples, from 35.9 ± 5 to 47.5 ± 6.6 µSv y^{-1} for pork samples and from 19.8 ± 2.7 to 32.3 ± 4.4 µSv y^{-1} for chicken. The contributions of the radionuclides to the annual effective dose due to meat consumption are the following: 76.20% from ^{226}Ra, 20.06% from ^{40}K, 2.69% from ^{210}Po, 0.74% from ^{210}Pb, 0.18% from ^{232}Th, 0.11% from ^{238}U. In this work, the annual effective dose due to intake of ^{40}K from chicken sample is 3.17±0.54 µSv y^{-1}, which is comparable with the value of 3.8 µSv y^{-1} reported by [Chae J. S., 2016]. Although the highest effective dose per kg was calculated for fish samples, the highest annual effective dose was found for pork samples. This might be due to the annual consumption rate of pork meat in Romania, which is 5.5 times higher than the fish consumption rate.

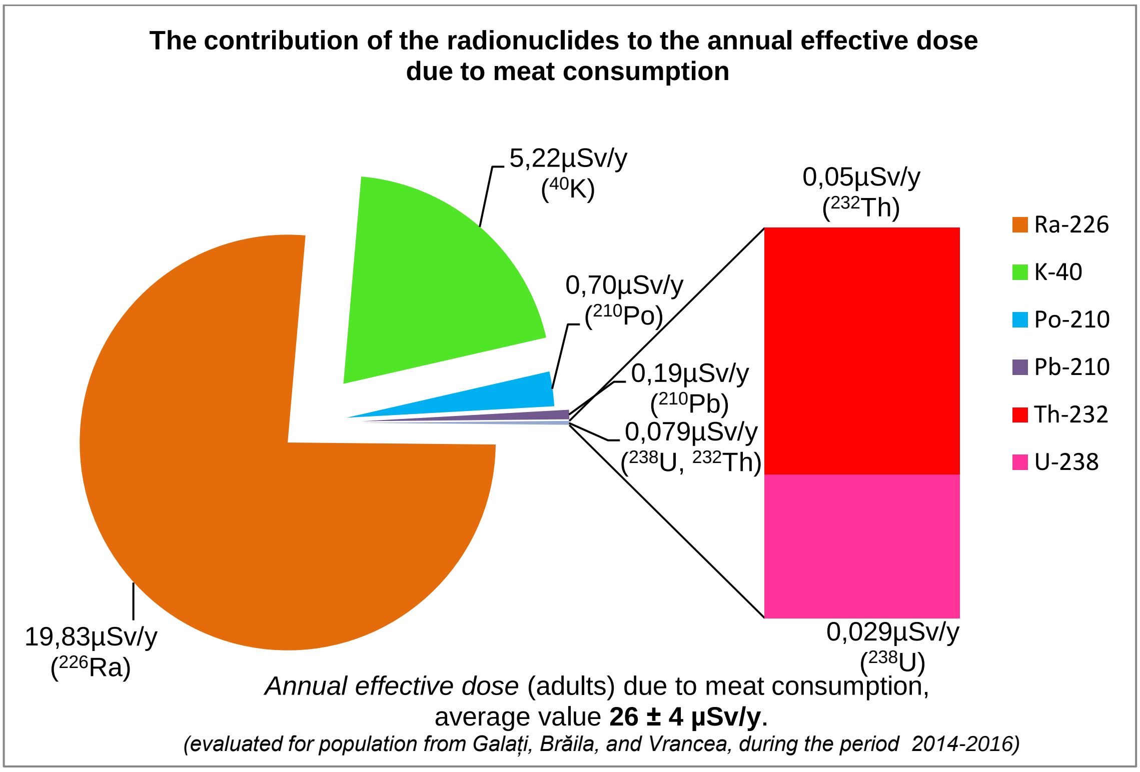


Figure 4.22 – *The contribution of the radionuclidesr ^{210}Po, ^{210}Pb, ^{238}U, ^{232}Th, ^{226}Ra și ^{40}K. to the annual effective dose due to meat consumption evaluated for the population from Galați, Brăila, and Vrancea counties, during the period 2014-2016*

The value of the annual effective dose due to the ingestion of radionuclides through the consumption of meat, evaluated in the present study, is 26 ± 4 µSv $year^{-1}$ - for adults. This is lower than the annual effective dose due to the ingestion of radionuclides from the uranium and thorium series, derived from adult food consumption (food and water), evaluated by UNSCEAR at: 110 µSv $year^{-1}$, [UNSCEAR, 2000] . In order to reach this value of 110 µSv $year^{-1}$ an annual meat consumption of 68.3 kg of fish, 84.64 kg of pork and 91.76 kg of chicken meat would be required, which would mean an annual increase in consumption of meat on average 12.5 times - for fish, 2.7 times - for pork and 4 times - for chicken, compared to the average consumption in Romania.

The contributions of radionuclides to the annual effective dose due to meat consumption are shown in *Figure 4.22*. Using alpha spectrometry, instead of gamma spectrometry, for a better sensitivity of ^{226}Ra measurements, the contribution of radionuclides to the annual effective dose could be significantly changed.

Using for the value for the detriment adjusted nominal risk coefficient for cancer and heritable effects published by International Commission on Radiological Protection (ICPR), which is 5.7×10^{-2} Sv^{-1} [ICRP, 2007], the statistical values for the life-time in Galati (75.22 years), Braila (74.87 years) and Vrancea (75.64 years) counties [http://statistici.insse.ro accessed: 30 November 2017], and the annual effective doses calculated, the lifetime cancer risk from meat consumption was estimated. The the lifetime cancer risk values ranged from 1.44 x 10^{-5} ÷ 20.37 x 10^{-5}, with an average of 11,14 x10^{-5}. This value is much lower than the total detriment from all natural radiation sources published by UNSCEAR (totaling 2.4 mSv y^{-1} annual dose) [ICRP, 2007], which average value was calculated as being 11.27×10^{-3}, thus demonstrating that the contribution to the annual effective dose due to the meat consumption is insignificant. This value is negligible compared to the detriment due to all natural sources of 2.4 mSv y^{-1} [Unscear, 2000], namely 10.27 x 10^{-3}, evaluated for the region of Galați, Brăila and Vrancea counties, between 2014-2016.

This is the first detailed study for the determination of the concentration of natural radionuclides (^{210}Po, ^{210}Pb, ^{238}U, ^{232}Th, ^{226}Ra, ^{40}K) in meat samples and for assessment of the annual effective dose due to their consumption in the Galați, Brăila and Vrancea counties.The value for lifetime cancer risk due to meat consumption is much lower than its value due to natural radiation sources. The consumption of meat is not a risk from the radiologically point of view.

The assessment to the annual effective dose due to powder milk consumption for babies aged 0-12 months

Twenty-four samples samples of the most available types of powder milk among various brand names were collected from the pharmacies in Galati, during the period from 2016 to 2017. The gross alpha and beta activity, the concentration of ^{210}Po, ^{210}Pb, ^{238}U, ^{232}Th, ^{40}K were determined, Based on these determined radioactivity parameters, the effective dose due to consumption of powdered milk by children (0-12 months) was evaluated.

During the first year of life, the amount of milk powder administered daily is very varied. Supplementing the powdered milk diet for children (0-1 years old) is personalized, depending on the body mass, identified needs, natural nutrition intake. Following these, the determined radionuclide concentrations were reported per kg of milk powder.

The values of the determined radionuclide concentrations were compared with the data from the scientific literature. The concentration of ^{210}Po, the most radiotoxic natural radionuclide, determined in the present study, is in the range: 0.014÷0.052 Bq kg^{-1}, the average being 0.026 Bq kg^{-1}. For the concentration of ^{210}Po, in Slovenia, values were determined in the range 0.055÷0.467 Bq kg^{-1}, [Štrok M., 2011] and in the range: 0.162÷0.358 Bq kg^{-1}, [Trdin M., 2017]. In Italy an average value of 0.013 Bq kg^{-1} was determined [Meli M.A., 2014], and in India (Mumbai) the values in the range: 0.08÷0.23 Bq kg^{-1}, were reported [Prabhath R.K., 2015] .

The concentration values of ^{238}U varies, in the present study, in the range from 0.01 to 0.014 Bq kg^{-1}, while in Slovenia it varies in the range from 1.02 to 2.06 Bq kg^{-1} [Trdin M., 2017], in Malaysia these ranging from 0,20 to 3,02 Bq kg^{-1} [Mei-Wo-Yii, 2019], in Iraq ranging from 0,115 to 25 Bq kg^{-1} [Abbojassim A. A., 2015].

The ^{40}K radionuclide is found in the highest concentration, as expected, because ^{40}K is a primordial radionuclide widely dispersed in the medium and easily transferable in the food chain. The average value of the ^{40}K radionuclide, determined in the present study, is 44.64 Bq kg^{-1}, while in Malaysia the average value of 99.1 Bq kg^{-1} has been determined [Uwatse O. B., 2015]. The concentration of ^{232}Th was found to be 2.55 ± 2.48 Bq kg^{-1}, in milk powder produced in Malaysia [Uwatse O. B., 2015], compared to the average value determined in this study, respectively 0.012 ± 0.001 Bq kg^{-1}.In Iraq for concentration of ^{40}K was founded in interval 104-461 Bq kg^{-1} [Abbojassim A. A., 2015], in Nigeria this varies from 17,8 to 55,1 Bq kg^{-1} [Kolapo A. A., 2014], and in Arabia Sudită ^{40}K vries from 29 to 149 Bq kg^{-1}, [Alamoudi Z. M., 2013].

The average concentration of ^{232}Th in powder milk from Malaysia ranges from 0,20 to 2,83 Bq kg^{-1} [Mei-Wo-Yii, 2019], and from 0,40 to 1,86 Bq kg^{-1} [Priharti W., 2016, while in Iraq this ranges from 1,59 to 13,57 Bq kg^{-1} [Abbojassim A. A., 2015], and in Saudi Arabia was found the values in interval 0,56÷2,93 Bq kg^{-1} [Alamoudi Z. M., 2013]. compared to this study: 0,011÷0,014 Bq kg^{-1} (average value 012 ± 0,001 Bq kg^{-1}).

The cumulative effective dose resulted by summing the average values of the doses corresponding to the consumption of milk powder for the age category 0-6 months and 6-12 months. In the present study, the evaluation of the cumulative effective dose based on the determined radioactivity parameters led to values in the range from 194.20 to 325.61 μSv - for the first year of life, the average being 267.44 μSv $year^{-1}$.

The contribution of radionuclides ^{210}Po, ^{210}Pb, ^{238}U, ^{232}Th and ^{40}K to the annual effective dose due to the consumption of milk powder is shown in *Figure 4.24.*

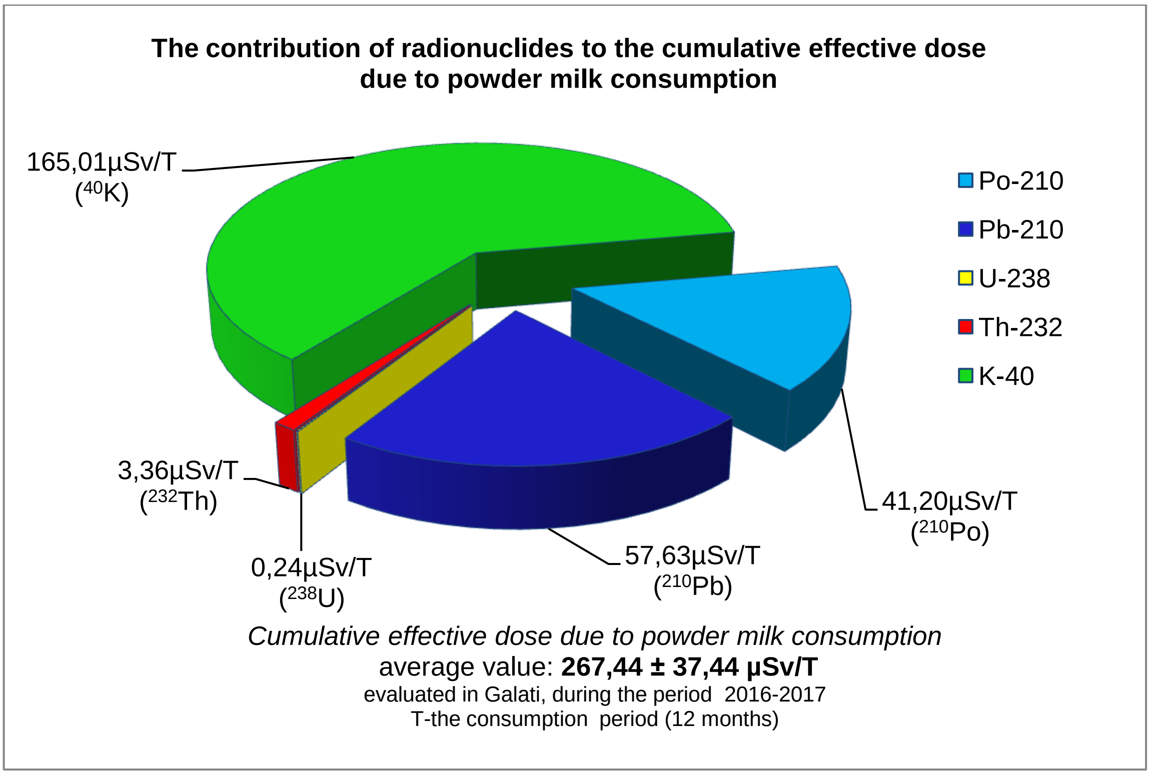


Figure 4.24 *– The contribution of the radionuclides ^{210}Po, ^{210}Pb, ^{238}U, ^{232}Th, ^{40}K, to cumulative effective dose due to powder milk consumption, evaluated in Galati county, during the period 2016-2017*

Taking into consideration the fact that, in order to consume powder milk, it is reconstituted by the addition of water, the assessment of the cumulative effective dose must also take into account the contribution of the natural radionuclides from the water used to prepare milk.

Based on the concentrations determined of rdionuclides, in the present study, the annual effective dose received by the population in the age category below 1 year was evaluated, at the average value of 72.3 µSv year^{-1} - in Galați county, in the period of 2015. Thus the average value of the cumulative effective dose due to the ingestion of radionuclides from both the powdered milk and the drinking water used for its reconstitution, is estimated at 339.74 µSv year^{-1}.

The contribution to the cumulative effective dose due to the ingestion of radionuclides through the consumption of powder milk increases in the order: ^{238}U (0.09%), ^{232}Th (1.25%), ^{210}Po (15.40%), ^{210}Pb (21.54%), ^{40}K (61.70%). The cumulative effective dose due to the ingestion of radionuclides ^{238}U, ^{232}Th, ^{210}Po, ^{210}Pb, received by the consumption of powdered milk, is evaluated at 102.43 µSv year^{-1}, comparable to 110 µSv year^{-1} - estimated by UNSCEAR, as the annual effective dose due to the ingestion of radionuclides from the uranium and thorium series.

4. CONCLUSIONS, ORIGINAL CONTRIBUTIONS, AND PERSPECTIVES OF RESEARCH

Conclusions

- ✓ The determination of the radioactivity parameters in the analyzed drinking water, from Galați, Brăila, Vrancea counties, between 2014 and 2018, revealed that the gross alpha and gross beta activity depends more on the geological structure of the area from which they originate and less of the technological processes for the production of drinking water.

- ✓ Evaluation of the annual effective due to drinkink water consumption, assuming the gross alpha, and gross beta activity, as a concentration of ^{210}Po, and ^{210}Pb/^{228}Ra, respectively, leads an over-assessment of the annual effective dose.dose. Nevertheless this method is fast and cover the maximum risck regarding to exposure of the population to ionising radiations by water consumption.

- ✓ For the region and period studied, the highest contribution to the annual effective dose due to the ingestion of natural radionuclides (^{210}Po, ^{210}Pb, ^{238}U, ^{232}Th, ^{226}Ra) through drinking water consumption, for adults, belongs to the radionuclide ^{210}Po, while the smallest contribution is due to radionuclide ^{238}U.

- ✓ The consumption of mineral water introduces in the human body, in the case of adults, a dose approximately two times lower than that introduced by drinking water consumption. The largest contribution to annual effective dose due to mineral water consumption belongs to radionuclide ^{226}Ra.

- ✓ The contribution of water and food consumption to the annual effective dose, evaluated for the adult population from Galați, Brăila and Vrancea counties, between 2013-2017, increases within the analyzed products as follows: mineral water (5.34 ± 0.81 µSv year^{-1}), drinking water (10.4 ± 1.3 µSv year^{-1}), bread (19.1 ± 3.4 µSv year^{-1}), pasteurized milk (29.4 ± 3.2 µSv year^{-1}), meat (26 ± 4 µSv year^{-1}), menu (483.5 ± 71.6 µSv year^{-1}).

- ✓ The contribution to the cumulative effective dose due to the ingestion of radionuclides through the consumption of milk powder increases in the order: ^{238}U (0.09%), ^{232}Th (1.25%), ^{210}Po (15.40%), ^{210}Pb (21.54%), ^{40}K (61.70%). The cumulative effective dose due to the ingestion of radionuclides ^{238}U, ^{232}Th, ^{210}Po, ^{210}Pb, received by the consumption of powdered milk, is evaluated at 102.43 µSv year^{-1}, comparable to 110 µSv year^{-1}- estimated by UNSCEAR, as the annual effective dose due to the ingestion of radionuclides from the uranium and thorium series.

- ✓ The evaluation of the annual effective dose, based on the radionuclide concentration (^{210}Po, ^{210}Pb, ^{226}Ra, ^{238}U) for the population of Galați, Brăila and Vrancea counties, from 2013-2018, due to the consumption of water and food, led to values below 100 µSv / year - in the case of water (recommended value not to be exceeded according to the European Directive 51/2013), respectively 110 µSv year^{-1}- for food (value estimated by UNSCEAR as the effective annual dose due to the ingestion of radionuclides from the uranium and thorium series).

✓ Measurements of the concentration of ^{222}Rn in the air of the public buildings and dwelling, in Galați county, revealed values well below the warning value, ie of 300 Bq / m3 - according to the European Directive 59/2013.

✓ Following the determinations performed in this thesis, it is concluded that drinking water, mineral water and food, from the territories of Galați, Brăila and Vrancea counties are safe in terms of natural radioactive content, contributing to the effective annual dose of the population below the limits established by the actually legislation.

Original contributions

This is the first integrated study to quantify the population's exposure to natural ionising radiation due to ingestion of radionuclides from water and foodstuffs consumption, and assessing the detriment to health correspondent to this consumption. The study was performed for a population of 1.15 million people, which means 5,84% from population of Romania.

The novelty aspects of this thesis:

➢ *establishing a significant data base for a period of 4 years, regarding the natural radioactive content of drinking water, mineral water and foodstuffs, used on an area of 14085 km2 (http://statistici.insse.ro) from the territory of Romania, representing the region of 3 counties (Galați, Brăila, Vrancea), populated by a number of people from 1.15 million people (http://statistici.insse.ro:8077/tempo-online/#/pages/tables/insse-table, accessed at dated 13.07.2019),*

➢ *comparative study, consistent from the point of view of the data series, evaluating the annual effective dose due to drinking water consumption, on the one hand, based on gross alpha and gross beta activity, and on the other hand, on the basis of radionuclide concentration with high conversion factor from specific activity to dose (^{210}Po, ^{210}Pb, ^{nat}U, ^{nat}Th, ^{226}Ra),*

➢ *construction of maps regarding the spatial distribution of gross alpha and gross beta activity in drinking water, as well as of the annual effective dose due to drinking water consumption, in Galați county in 2014-2017 and Vrancea county in 2015-2018;*

➢ *assessment of the level of natural radioactive content of drinking water for three cities in Romania: Galați, Brăila and Vrancea, between 2014-2018, through determinations of gross alpha activity, gross beta activity and concentration of ^{210}Po, ^{210}Pb, ^{nat}U, ^{nat}Th, ^{226}Ra ;*

➢ *evaluation of the natural radioactive content of foodstuffs such as: bread, meat, pasteurized milk, menu for preschoolers, powdered milk for children from 0-12 months age group, from Galați, Brăila and Vrancea counties, between 2014-2018, by determining concentration of ^{210}Po, ^{210}Pb, ^{nat}U, ^{nat}Th, ^{226}Ra and gross alpha and gross beta activity;*

➢ *evaluation of the annual effective dose for the population of Galați, Brăila and Vrancea counties, due to the ingestion of radionuclides (^{210}Po, ^{210}Pb, ^{nat}U, ^{nat}Th, ^{226}Ra) through the consumption of drinking water and foodstuffs in the period 2014-2018,*

- *evaluation of the detriment due to ionisin radiations through water and foodstuffs consumption*

- *evaluation of the annual effective dose due to the inhalation of radon from the air of public buildings and dwellings in Galați county*

Research directions

This study opens the following research directions:

- *epidemiological studies regarding the association of radionuclides in drinking water and food with diseases related to the ingestion of radionuclides through their consumption,*
- *studies on the correlation between the radioactive content of the drilling water and the geological composition of the soils from which they originate,*
- *studies on the content of artificial radionuclides in water and food and the evaluation of the effective dose, respectively of the detriment received due to their ingestion,*
- *studies regarding the concentration of radionuclide* ^{228}Ra *in drinking water as well as in food components and completing the evaluation of the annual effective dose due to the ingestion of natural radionuclides through the consumption of drinking water, mineral water and food (evaluated in the present study) with the contribution of this radionuclide,*
- *the study of the exposure of the population to ionizing radiation due to other sources of ionising radiations (eg sources of exposure to ionizing radiation through medical exposures),*
- *studies on the radioactive content of building materials,*
- *studies regarding the improvement of the radiochemical methods of separation in order to determine the content of natural radionuclides in water and foodstuffs (eg directions of improvement: shortening the time of development, increasing specificity, reducing the need for radioactive tracers, etc.), standardizing methods.*

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