UNIVERSITATEA LUCIAN BLAGA DIN SIBIU

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DOCTORAL THESIS

CONTRIBUTIONS TO THE DEVELOPMENT OF A MULTIVARIATE SPATIO-TEMPORAL MODEL REGARDING THE RADIOACTIVITY, RADON-222 AND PHYSICAL CHEMICAL PARAMETERS IN DRINKING WATER FROM SIBIU AND ALBA COUNTIES

-SUMMARY-

Ph.D. student:
Chem. CECILIA IONELA ȘITOIANU (married TĂBAN)

Ph.D. supervisor:
Prof. univ. dr. habil. SIMONA OANCEA

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List of abbreviations

MDA - Minimum Detectable Activity
DNA - Deoxyribonucleic acid
Al - Aluminum
Bq - Bequerel
Cd - Cadmium
MAC - Maximum Allowable Concentration
NCCNA - National Commission for the Control of Nuclear Activities
NCMRCE – National Center for Monitoring Risks in the Community Environment
AAED - Average Annual Effective Dose
AED - Annual Effective Dose
TERD - Total Effective Reference Dose
PHD - Public Health Directorate
EDTA - ethylenediaminetetraacetic acid
Fe – Iron
^{137}**Cs**-cesium-137
^{90}**Sr**-strontium-90
^{226}**Ra**-radium-226
^{222}**Rn**-radon-222
CF - Concentration Factor
CF – Conversion Factor
GeHP - Hyper Pure Germanium
GRP - Geogenic Radon Potential
GLMM - Generalized Linear Mixed Model
GLM - Generalized Linear Model
Gy - Gray
HDPE - High Density Polyethylene
IAEA - International Atomic Energy Agency
IARC - International Agency for Research on Cancer
ICRP - International Commission on Radiological Protection
IFIN HH - National Institute for Research and Development in Physics and Nuclear Engineering "Horia Hulubei".
NIPH - National Institute of Public Health
ISO - International Organization for Standardization
J – Joule
LDPE - Low Density Polyethylene
RIL - Radiation Hygiene Laboratory
MDA - Minimum Detectable Activity
Mn - Manganese
mCi – miliCurie
µCi - microCurie
RM - Reference Material
mSv - miliSievert
MH - Ministry of Health
NORM - Radioactive Material Present in Nature
WHO - World Health Organization
GEO - Government Emergency Ordinance
Pb - Lead
MCA - Main Component Analysis
pCi - picoCurie
PE - polyethylene
PET - Polyethylene Terephthalate

PLBP - Polylactic Biopolymer
PP - polypropylene
RDA - Redundancy Analysis
RENAR – Romanian Accreditation Association
SI - International System
Sv - Sievert
EU - European Union
UNSCEAR - The United Nations Scientific Committee on the Effects of Atomic Radiation
US EPA - United States Environmental Protection Agency
DWSA - Drinking Water Supply Areas

Thesis Summary

1. Introduction

A special interest is given by researchers in the field, but also by the staff responsible for the management of drinking water sources, to certain radionuclides and their degradation products, because these pollutants generated from natural or anthropogenic sources can contaminate water (Qin-Hong et al., 2010). Among the radionuclides, radon, Rn-222, is a radioactive gas from the decay of radium, the latter being the degradation product of uranium, U-238. Research studies have reported evidence of the carcinogenicity of the radioactive gas Rn-222 (Rafiquei et al., 2012), which is mostly responsible for internal exposure in humans, according to the report of the *International Commission on Radiation Protection*. Drinking water sources in groundwater may contain radon, which is a toxic radioactive gas (WHO, 2017). Of great concern is the presence of Rn-222 in water, which has led to an increase of studies aiming to assess its levels in different water sources during the past two decades (Abdallah et al., 2007). In deep waters, the values of this radioactive element radon-222 are higher compared to the values in surface waters, and therefore more thorough research is required (Byong Wook Cho et al., 2007).

In recent years, radioactivity found in groundwater became a serious problem worldwide (Gado et al., 2018). Radionuclides dissolved in water emit particles (alpha, beta, and gamma), which gradually affect all living tissues (Alam et al., 1999; Gruber et al., 2009). All these radioactive substances have devastating effects over time, causing various diseases including cancer.

Heavy metals may occur naturally in the environment. Some are biologically essential, i.e. they play an important role in biochemical metabolism. However, the bioaccumulation of these metals can disrupt the function of vital organs such as liver, kidneys, heart, and many studies are indicating carcinogenicity caused by some heavy metals (Taiwo et al., 2018; Mohammadi et al., 2019). Transport of heavy metals in the environment can be natural (Chen et al., 2018a, b; Eid et al., 2018) and anthropogenic (Zhang et al., 2019), such as heavy metals released from mining activities, industrial waste, exhaust from vehicles, and agricultural activities. Monitoring radioactivity and physicochemical parameters in drinking water is essential to ensure safe and healthy water for the population's consumption. It protects public health, ensures regulatory compliance, and prevents damage to water supply infrastructure. It is also very important to implement effective monitoring and control systems to guarantee the quality of drinking water.

In this context, the purpose of this doctoral thesis is to evaluate the important radiological parameters of water samples collected from different drinking water sources in two studied counties, namely Sibiu and Alba, their correlation with physicochemical parameters, and to develop a multivariate spatio-temporal mathematical model. This purpose will lead to measures to eliminate risks of contamination or exposure to radioactive isotopes such as radon and to control risk of radioactive levels that can be present in different sources of drinking water and can cause serious health problems, including cancer.

2. Descriptive research on the evaluation of drinking water quality in the light of the investigation of physicochemical and radiological parameters

It has been shown that certain chemical contaminants can cause adverse effects on human health as a result of exposure through drinking water. The likelihood of a given chemical occurring in significant concentrations in a given setting must be assessed on a case-by-case basis (WHO, 2022).

The monitoring of the quality of drinking water distributed in centralized systems of large and small supply areas is carried out with the aim of improving the quality and safety of drinking water in Romania by detecting risks to the health of the population and protecting human health from the harmful effects of pollution of water intended for human consumption, by ensuring the quality of water, hygienic and clean, and with the aim of expanding the access of the population in the future.

2.1 National situation regarding the quality of drinking water distributed in a centralized system in small and large supply areas

Figure 1 shows the situation regarding the number of large drinking water supply areas (i.e. areas with more than 5000 consumers or with a drinking water supply of more than 1000 m^3/day) in the municipality of Bucharest and in 41 counties in Romania, in the period 2017-2021.

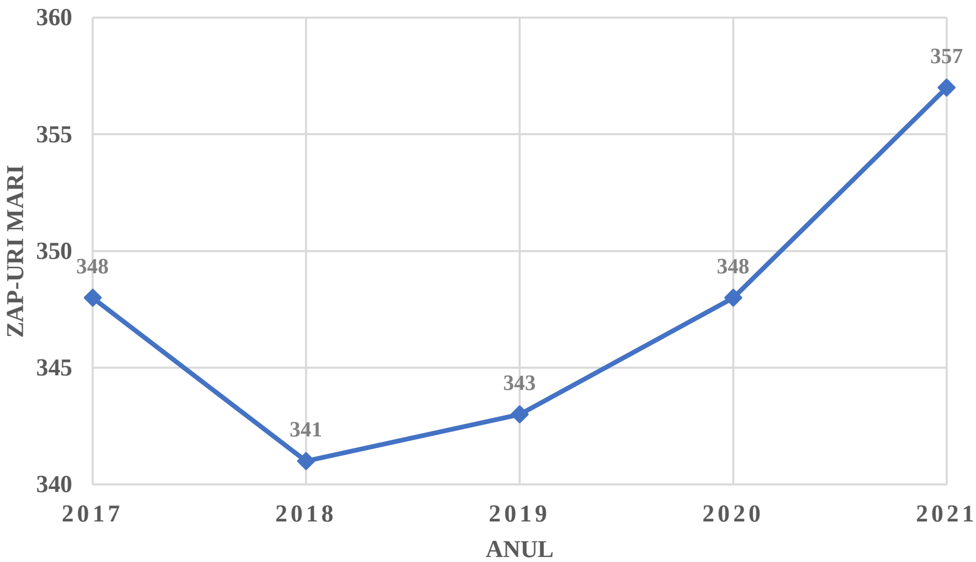


Figure 1. The situation regarding the number of large drinking water supply areas (DWSA) in the 41 counties and the Municipality of Bucharest in Romania, in the period 2017-2021.

By screening the quality of well water and artesian water or springs for public use, the physicochemical parameters in the 41 counties and the municipality of Bucharest are also monitored. For example, in the period 2017-2019, reporting sheets of physicochemical indicators were recorded from 39 counties out of the 41 of Romania and the municipality of Bucharest, and in 2020 and 2021 there were reporting sheets from only 37 counties and 38 counties respectively, in which the quality of drinking water intended for human consumption from public wells and springs was analyzed, resulting the statistical situation illustrated in figure 2.

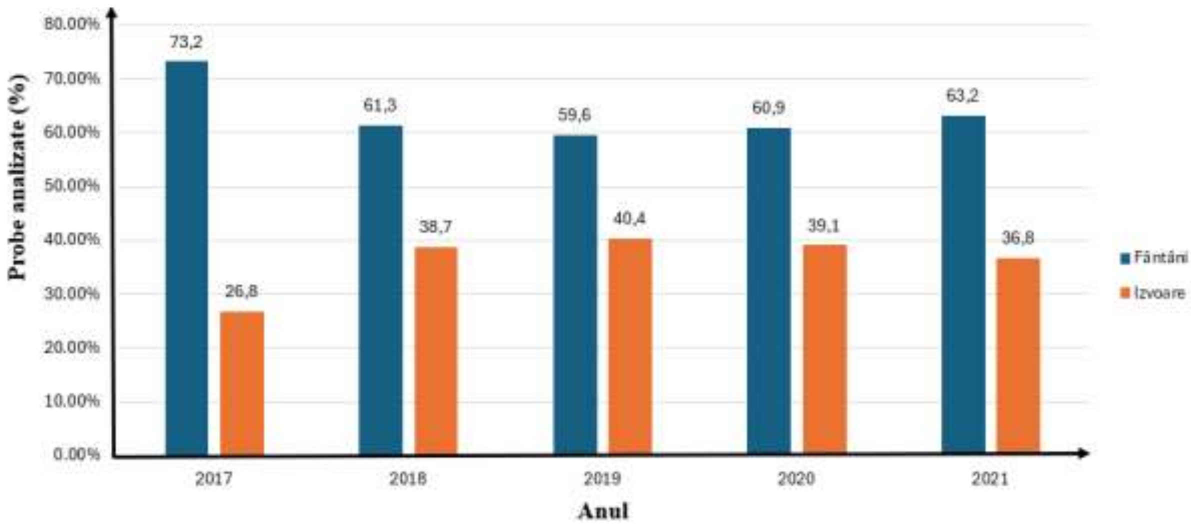


Figure 2. The share of drinking water analyses from wells and springs within the period 2017-2021, in Romania

At national level in the period 2017-2021, the exceeding of the maximum allowable concentration MAC of the physicochemical parameters of drinking water from public wells and springs, was found for nitrates, iron, ammonium, manganese, and turbidity, in different percentages of the total number of analyses found non-compliant (CNMRMC, 2017-2021); there are also analyses that were below the MAC for the parameters nitrates and ammonium in certain counties, for the same period of time (2017-2021).

The percentages of unsatisfactory results following the analyses reported by local PHDs for several metals in small drinking water supply areas (CNMRMC, 2017-2021) were as follow: manganese from 12.67% in 2017 to 15.15% in 2021, and iron from 8.81% in 2017 to 11.44% in 2021.

Table 1 shows the average values of alpha, global beta and ^{222}Rn activity concentrations in the period 2017-2021, at national level (CNMRMC, 2017-2021).

Table 1. The average values of global alpha and beta radioactivity, as well as the content of ^{222}Rn for water samples, from small and large supply areas in 36 counties and the municipality of Bucharest, 2017-2021.

Item No.	**Year**	**Global Alpha Activity (Bq/L)**	**Global beta activity (Bq/L)**	**^{222}Rn (Bq/L)**
1.	2017	0.0250	0.1080	5.640
2.	2018	0.0216	0.1143	1.621
3.	2019	0.0250	0.1100	1.650
4.	2020	0.0300	0.1000	4.500
5.	2021	0.0300	0.1080	5.000
Reference value (Law 301/2015)		**0.1**	**1**	**100**

The most accepted protocol used as a first step in the radiological characterization of drinking water is the determination of global alpha and beta activity (Todorovic et al., 2012; Jobbagy et al., 2012) in accordance with standards ISO 9696:2018 and ISO 9697:2019 for non-saline water. If the concentration of global alpha activity is < 0.1 Bq/L, and the global beta activity < 1 Bq/L, respectively, the total annual effective dose due to drinking water ingestion is estimated to be lower than the reference value of 0.1 mSv/year (Law 301/2015).

2.2 Research on the content of radioactive elements ^{137}Cs, ^{90}Sr and ^{226}Ra in drinking water in the counties of Sibiu and Alba

Drinking water contains natural radionuclides such as uranium-radium (^{238}U-^{226}Ra) and thorium (^{232}Th) of the decay series, with their degradation products and potassium (^{40}K), as well as artificial radionuclides (^{137}Cs, ^{134}Cs, ^{90}Sr, etc.) that come from atmospheric nuclear weapons testing and nuclear reactor accidents. Following radioactive fallout after the Chernobyl nuclear reactor accident in 1986, the radionuclide ^{137}Cs ($t_{1/2}$ = 30.07 years) was widely dispersed in the Turkish environment (Turhan et al., 2012). These radionuclides could pose a risk to human health (WHO, 2011). Low doses caused by the ingestion of these radionuclides from drinking water may increase the radiological risk of longer-term effects.

Therefore, the determination of concentration levels of natural and artificial radionuclides in drinking water is an important factor for public health studies, which allow the assessment of the population's exposure to radiation through water consumption.

The main objective of this study was to evaluate the levels of ^{137}Cs, ^{90}Sr and ^{226}Ra in the drinking water samples collected from areas located in the counties of Sibiu and Alba, which are important in monitoring the radiological hazards caused by the consumption of these waters and the possible changes in the radioactivity of the environment following specific actions, nuclear, industrial, or other human activities with a negative impact on the environment.

2.2.1 Results and discussions

The experimental data on the levels of radionuclides ^{137}Cs, ^{90}Sr and ^{226}Ra in drinking water that were processed and interpreted in this study, were extracted from laboratory reports based on their own analytical determinations, over an interval of 6 years (2000 – 2005), the water samples being collected from various regional points of the two counties, Sibiu and Alba.

From the study of the level of beta emitting radionuclides ^{137}Cs and ^{90}Sr carried out over 4 years (2000-2003) detected in drinking water samples from the county of Sibiu, it is seen that the minimum and maximum value of the ^{137}Cs concentration was 0.0010 Bq/L (mainly at the regional points of Tălmaciu, Cristian, Cisnădie) and respectively 0.0040 Bq/L (mainly at the regional points of Mediaș, Dumbrăveni). In the case of the ^{90}Sr concentration, the highest values (0.0040 Bq/L) were obtained mainly from the samples from Mediaș, and the lowest values (0.0010 Bq/L) mainly from the drinking water samples from Cristian, Tălmaciu, Dumbrăveni and Cisnădie.

For the alpha emitting radionuclide ^{226}Ra evaluated in the drinking water samples from the county of Sibiu for 6 years (2000-2005), it is seen that the minimum and maximum value was 0.0017 Bq/L in 2000 in the Mediaș drinking water sample, respectively 0.0740 Bq/L in 2005 in the Miercurea Sibiului water sample.

From the study of the level of beta emitting radionuclides ^{137}Cs and ^{90}Sr carried out over 4 years (2000-2003) detected in drinking water samples from the county of Alba, it is seen that the minimum and maximum value of the ^{137}Cs concentration was 0.0010 Bq/L (mainly at the regional points of Alba, Arieșeni and Zlatna) and respectively 0.0120 Bq/L and 0.0050 Bq/L at the regional points of Alba and Sebeș. In the case of the concentration of ^{90}Sr, the highest values, respectively 0.0080 Bq/L, were obtained mainly from the drinking water sample in Câmpeni (2001), and the lowest values, respectively 0.0010 Bq/L, mainly from the drinking water samples of Alba, Arieșeni, and Zlatna.

For the alpha emitting radionuclide ^{226}Ra determined in the drinking water samples from the county of Alba for 6 years (2000-2005), it is seen that the minimum and maximum value was 0.0010 Bq/L in 2001 in the Zlatna drinking water sample, respectively 0.0070 Bq/L in 2003 also in the Zlatna drinking water sample.

For Sibiu County, the values of radionuclide concentrations are much lower than those recorded in Alba County, but there are relatively high values of ^{226}Ra (0.0440 Bq/L) in 2004 in the Apoldu de Sus drinking water sample and 0.0740 Bq/L in the Miercurea Sibiului drinking water sample in 2005.

3. Experimental research on the physicochemical parameters and radioactivity indicators of drinking water from different sources in the county of Sibiu. Development of a multivariate model on drinking water quality.

The quality of drinking water in Romania is researched through the national program for monitoring environmental and occupational risk factors. Effective monitoring of drinking water is extremely necessary to reduce the impact of water pollution on human health, especially for the prevention of cancer, infectious diseases, and gastrointestinal diseases (Lin, Yang, & Xu, 2022). While infectious diseases caused by inadequate water consumption are well documented, with 34% attributable to inadequate drinking water (World Health Organization, 2016), the impact of water radioactivity and chemical contamination on health remains a cause for concern.

Tools such as multivariate statistical techniques, water quality identification index, positive matrix factorization or the Soil Water Assessment Tool have been applied to examine the spatio-temporal variation of water quality, to determine major sources of pollution in rivers and to optimize environmental risk factor management practices (Shrestha and Kazama, 2007; Wang et al., 2015; Geng et al., 2019). However, multi-level models are suggested to be the default statistical approach in ecological research, as they better address dataset analyses when traditional statistical assumptions are not met (Schreiber et al., 2022).

The main objectives of this research consisted in the evaluation of the global alpha and beta radioactivity, of the radon-222 content, and of the mainly physicochemical parameters, with the development of a multivariate mathematical model that would provide a dynamic model of the drinking water quality for a geographically defined area: the county of Sibiu, regarding the physicochemical and radiological indicators, which can be useful for establishing the short-term dynamics and continuous monitoring for the operational and audit management.

3.1 Materials and methods

The analyses were carried out on a number of 65 drinking water samples from the county of SIBIU

- **Global alpha and beta radioactivity method:** according to SR ISO 9696/2018 and SR ISO 9697/2019 respectively;
- **Radon 222 concentration method:** according to SR EN ISO 13164-3:2020 standard;
- **Methods of physicochemical parameters:** according to the standards of each parameter.
- **Development of a mathematical model**: generalized linear model, generalized linear mixed models with gamma distribution and multivariate linear redundancy analysis.

3.2 Results and discussions

For the study and research proposed in this chapter, drinking water samples were evaluated, collected annually from 13 locations illustrated in Figure 3, in the period January - November, between 2017 and 2021. The diversity of water sources in the supply areas and the predetermined periodicity of sampling were considered eligible criteria for the present study. In this regard, the following sources of drinking water were selected, as can be seen in Figure 3.

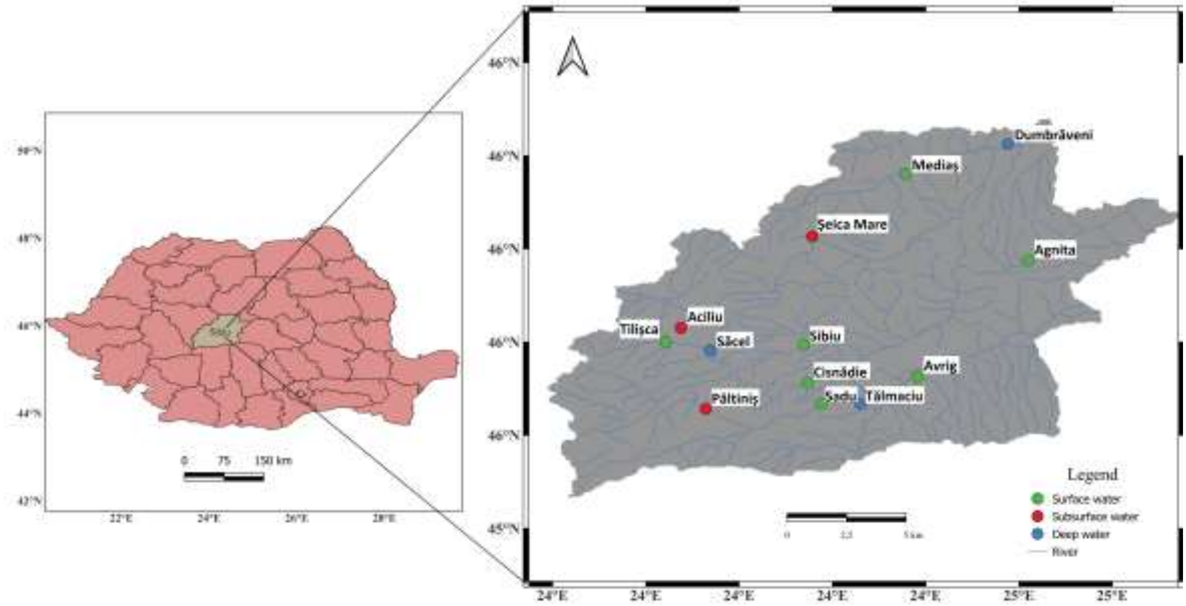


Figure 3. Location of sample collection points and water sources

3.2.1 Evaluation of the radioactivity indicators of drinking water samples from sources in the county of Sibiu.

Results: the global alpha activity in drinking water over a period of 5 years (65 determinations) ranged from 0.0011 to 0.0842 Bq/L, with an average value of 0.0101 Bq/L. The highest global alpha activity was recorded in the drinking water collected from Tilișca (surface water) and the lowest value of the global alpha activity in the drinking water from Sibiu (surface water) in September 2018; the global beta activity in drinking water over a period of 5 years (65 determinations) ranged between 0.0296 Bq/L and 0.3980 Bq/L, with an average value of 0.0986 Bq/L, the highest global beta activity being recorded in the Tilișca drinking water in September 2017 and the lowest value being recorded in the Păltiniș drinking water (from the captured springs) in June 2018. The highest total global alpha and beta activity was recorded in the Tilișca drinking water during September 2017, and this may be the result of the geological composition, but also of certain precipitations, through the runoff of rainwater into rivers and lakes.

The results on global alpha and beta radioactivity, respectively, in this study, presented comparatively with those of other published studies, are described in Table 2.

Table 2. Comparison of the results on global alpha and beta activity, respectively, in this study with those published in other studies.

Item No.	Water Sample	Region/Country	Global alpha activity (average) (Bq/L)	Global beta activity (media) (Bq/L)	Bibliographic references
1.	Surface and Depth Water	Sibiu/Romania	0.0011-0.0842 (0.0101)	0.0296-0.3980 (0,0986)	**This study**
2.	Surface water	Adana/Turkey	0.0003-0.0229 (0.0096)	0.0189-0.2907 (0.086)	Degerlier et al., 2010
3.	Surface water	Tarkwa Goldmine /Ghana	0.008-0.017 (0.012)	0.063-0.374 (0137)	Faanu et al., 2010
4.	Surface water	Ebro River /Spain	(0.095 ± 0.004)	(0.213 ± 0.012)	Pujol and Sanchez, 2000
5.	Surface water	Fitina River /Turkey	0.0124-0.0662 (0.0326)	0.0279-0.1333 (0.0699)	Kucukomeroglu et al., 2008
6.	Groundwater	Kumasi/Ghana	0.0157-0.1427 (0,0407)	0.0893-0.4	Darko et al., 2014
7.	Groundwater	Adentan/Ghana	0.0049-0.086 (0.034)	0.026-1.867 (0.501)	Eric et al., 2013

According to Eric et al. (2013), the maximum value of global beta activity was 1.867 Bq/L, which means that the maximum limit recommended by the WHO of 1 Bq/L is exceeded. In general, most of the samples had the mean values of global alpha and beta activity lower than those established in the WHO guidelines.

Global alpha and beta activities generally serve as a mean of screening the quality of water samples from the radiological point of view. The WHO has recommended guidelines on global alpha and beta activity in drinking water with permissible values of 0.5 Bq/L and 1.0 Bq/L, respectively.

The comparison of the ^{222}Rn results of this study with those reported by other countries is presented in Table 3.

Table 3. Comparison of the concentration of ^{222}Rn in this study with that reported in published studies from other countries.

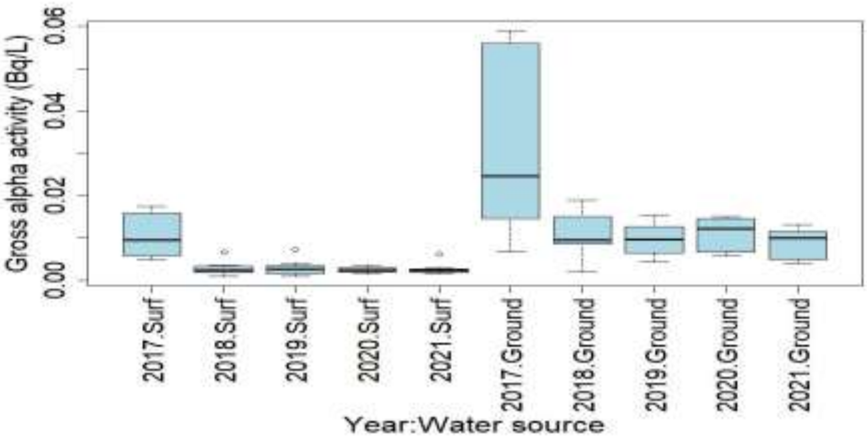
Crt. No.	Region/Country	Concentration of ^{222}Rn (Bq/L)		Bibliographic references
		Value min.-max.	Average Value	
1.	Sibiu, Romania	0.034 – 43.302	4.111	**This study**
2.	Punjab, India	1.4 – 5.3	3.5	Jakhu et al., 2020
2.	Kericho County, Kenya	4.6 – 22.5	12.4	Rotich et al., 2020
3.	Kurdistan, Iraq	1.1 – 10.3	6.8	Ezzulddin and Mansour, 2020
4.	Jerba Island, Tunisia	0 - 2860	867.0	Telahigue et al., 2018
5.	Rajasthan, India	12.5 – 862.0	113.0	Duggal et al., 2020
6.	Covilhas County, Portugal	2.0 - 1690	352.8	Inacio et al., 2017
7.	Zarand City, Iran	5.2 – 14.4	9.7	Blush et al., 2020
8.	Southwest, Nigeria	1.6 – 271.0	35.9	Ajibove et al., 2018
9.	Jazan, Saudi Arabia	1.6 – 3.82	2.0	El-Araby et al., 2019
10.	Spain, La Garrotxa region	0.8 – 26.0	11.4	Moreno et al., 2014

The average value of the concentration of ^{222}Rn of the 13 drinking water samples during the period of 2 years (26 determinations) was 4.111 Bq/L, and the lowest concentration of ^{222}Rn (0.0344 Bq/L) was recorded in the surface drinking water collected from the city of Avrig, while the highest (37.4770 Bq/L in November 2020, and 43.3025 Bq/L in April 2021), was recorded in the drinking water samples from Păltiniș, which is a mountain resort located at an altitude of 1440 m, 32 km north of the city of Sibiu. The values recorded in the deep drinking water of Păltiniș both in 2020 November and in 2021 in April, exceeded the US-EPA recommended reference value of 11.1 Bq/L, but did not exceed the WHO recommended limit of 100 Bq/L, being a lower average value compared to other studies such as Jerba Island/Tunisia (Telahigue et al., 2018), Covilhas County/Portugal (Inacio et al., 2017), Rajasthan/India (Duggal et al., 2020) that exceeded the US-EPA recommended level of 11.1 Bq/L, but also the WHO recommended limit of 100 Bq/L.

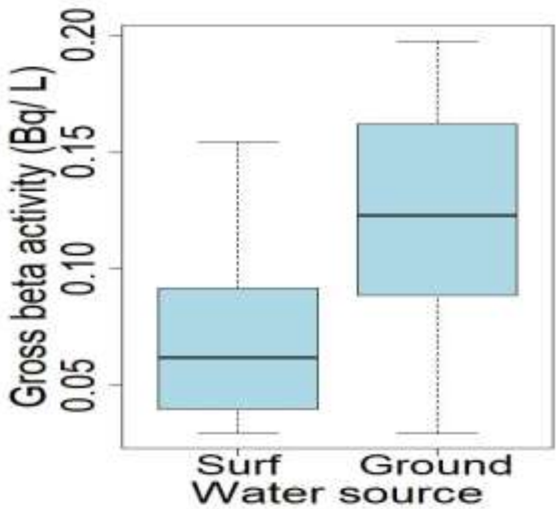
3.2.2 Statistical analysis and development of a mathematical model on radioactivity indicators of investigated drinking water samples (GLM, GLMM, RDA)

The main indicators studied of radioactivity of the water samples were: global alpha activity, global beta activity and radon-222 content.

Figures (4 a and 4 b) show the results of alpha and beta radioactivity by water sources, 2017-2021.



(a)



(b)

Figure 4. Box-and-whisker charts of global alpha radioactivity (**a**) and global beta radioactivity (**b**) by water sources, 2017-2021

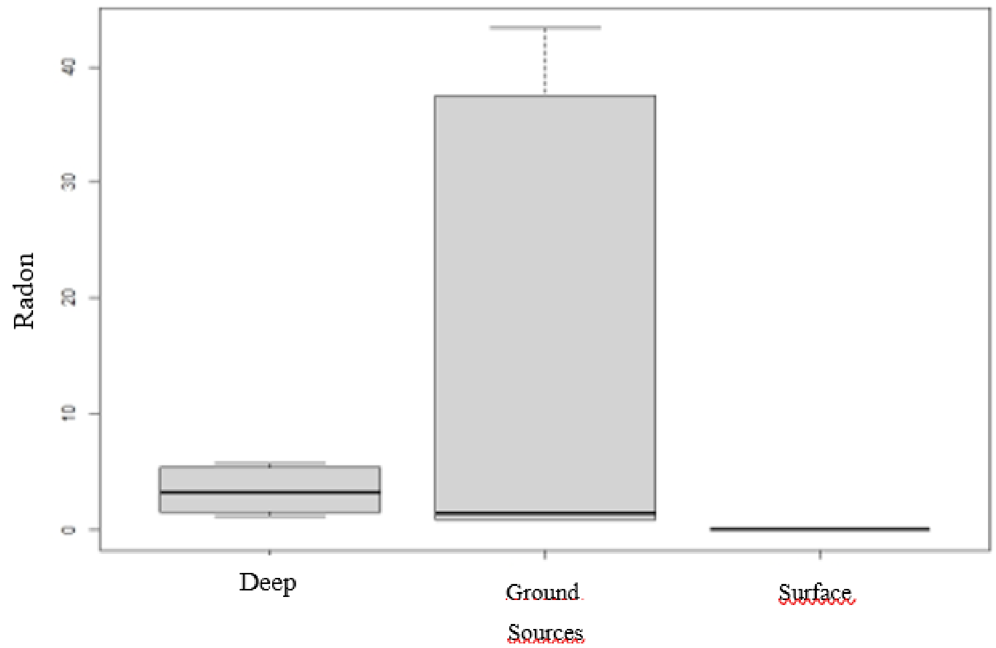


Figure 5. Box-and-whisker diagram of radon-222 concentration according to water sources in the county of Sibiu, between 2020 and 2021

The best model for these experimental data was GLMM with gamma error distribution. The variance of radon-222 concentration between localities was 1.54, and the average value for localities with surface water was 1.89. In localities with groundwater, the average concentration of radon-222 was 96% lower, and the difference was very significant ($t = -8.83$, $df = 22$, $p < 0.001$). The three types of water sources were the best predictors of radiation parameters (pseudo-$F = 10.8$, $p = 0.001$), explaining 48.4% (43.9% adjusted) of the variation in response variables.

The first constrained axis was the only significant one (pseudo-$F = 9.5$, $p = 0.001$), accounting for 93.7% of the explained variation. This axis was mainly defined by the difference between surface and groundwater sources. Along this axis, all radioactivity parameters increased from Surface to Depth and Ground (Figure 6).

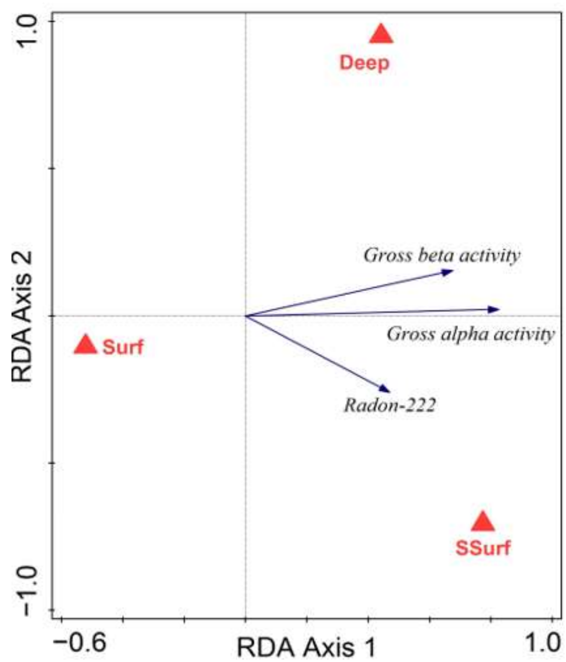


Figure 6. Graph of the redundancy analysis (RDA) of correlation of radiological parameters with the type of water sources. The first two constrained axes are illustrated, but the second axis is not significant

Global alpha and beta activities were higher in the DeepWater samples compared to the Surface ones (Figure 7a), while all three radioactivity parameters (global alpha and beta activity, radon-222) were significantly higher in the Groundwater samples compared to the Surface ones (Figure 7b).

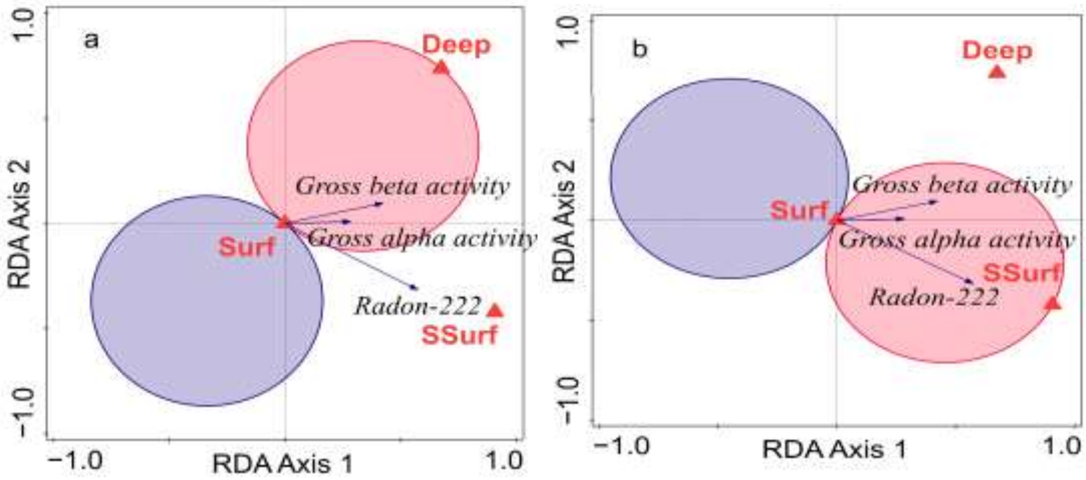


Figure 7. T-value graphs of radioactivity parameters for surface water in relation to source type: deep water (a) and groundwater (b). The pink circle delimits the ordering space for the significant positive response to the variable considered and the blue circle for the negative response.

However, the high level of variation explained by this model is mainly due to the particularly high values of radon-222 from Păltiniș, which has a groundwater source. If only global alpha and beta activity are considered, along with locality, the pattern was significant (pseudo-F = 2.1, p = 0.012), but the explained variation was smaller, at 32.7% (17.3% adjusted). The localities were scattered in the ordering space (Figure 8).

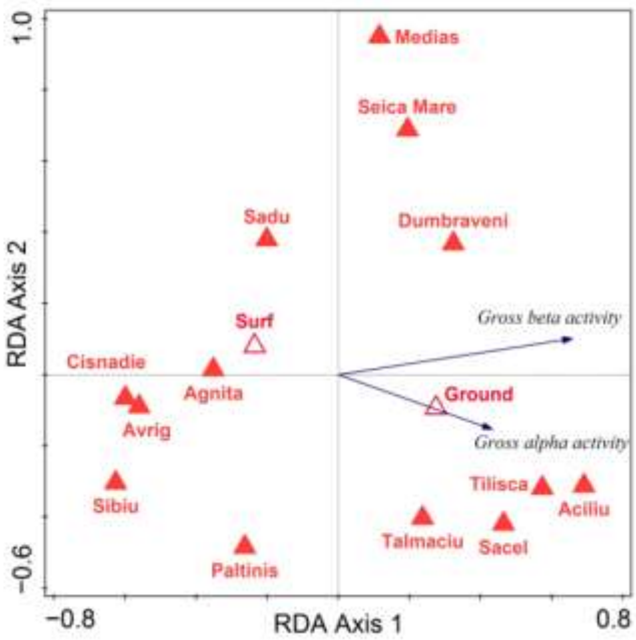


Figure 8. Redundancy analysis (RDA) graph of correlation of global alpha and beta activity with localities and surface and groundwater sources.

The first ordering axis, which was the only significant one (pseudo-F = 2, p = 0.012), was given by the difference between surface and groundwater sources, with global alpha and beta activities increasing in localities with groundwater samples. Sibiu and other larger cities (Cisnădie, Avrig) use surface water; therefore, the radiation level is the lowest, while some smaller localities (Aciliu, Tilișca, Săcel) use groundwater, except for Tilișca, and their radiation level is the highest (Figure 8). Tilişca, which uses surface water, is close to Aciliu, and the geology of the site may explain the particular radioactivity of this source.

3.2.3 Correlation analysis of the radioactivity of drinking water samples with their physicochemical characteristics

Of the 65 drinking water samples investigated in the present doctoral study, 16 samples exceeded the allowed values for the physicochemical indicators iron, ammonia, and residual free chlorine (table 4).

Table 4. Exceedances of the maximum allowable concentration (MAC) of some chemical contaminants depending on the water sources and locations investigated in the present study.

Location	Water source	Chemicals			Year of collection
		Ammonium	Residual chlorine	Iron	
Sadu	surface	x		x	2017, 2018, 2019
Tilişca				x	2018, 2021
Agnita				x	2020
Mediaș			x		2021
Sibiu				x	2018, 2019, 2021
Săcel	deep	x	x		2018
Şeica-Mare	ground	x	x		2017, 2021
Aciliu		x			2021

In terms of chemical composition, the drinking water sources in Avrig, Cisnădie, Dumbrăveni, Păltiniș and Tălmaciu meet all the quality criteria in the studied period 2017-2021.

Significant correlations were found between some physicochemical parameters (Table 5).

Table 5. Correlation coefficients between radioactivity and physicochemical parameters.

Parameter	Global Alpha Activity	Global beta activity	Fe	Cl	Cd	Mn	pH	Ammonium
Global beta activity	**0.709 (0.327)**							
Nitrates	**(0.344)**							
Fe		**-0.34**						
Mn		**(0.285)**						
pH	**-0.276**							
Ammonium				**0.303**				
Al					0.269	0.237		
Oxidability			0.238				-0.259	0.234
Conductivity	**0.497**	**0.527**					-0.263	
Turbidity		(0.257)	**0.338**			0.236		

Note: Only significant results ($p < 0.05$), bold, or marginally significant ($0.05 < p < 0.1$) are shown. The values given in parentheses are for the dates from which the outliers have been excluded.

The water sources (surface and ground) and the month of sample collection explained 29.1% (14.4%) of the variation in physicochemical parameters (pseudo-F = 2, p = 0.004). The first constrained axis was the only significant one (pseudo-F = 0.7; p = 0.002), accounting for 42.5% of the explained variation. This axis was mainly defined by the difference between surface and groundwater sources, but also by some monthly variations. Along this axis, turbidity and Fe recorded the highest values in surface water samples, while conductivity, nitrates, ammonia, and pH had the highest values in groundwater samples (Figure 9), with response to the water source being significant in Fe, turbidity, conductivity, and nitrates (Figure 10). During the sampling month, August was characterized by high values of Fe and turbidity, March by high pH and concentrations of Cl and ammonium, and January by high values of conductivity and nitrates, but also Cd and oxidability (Figure 9).

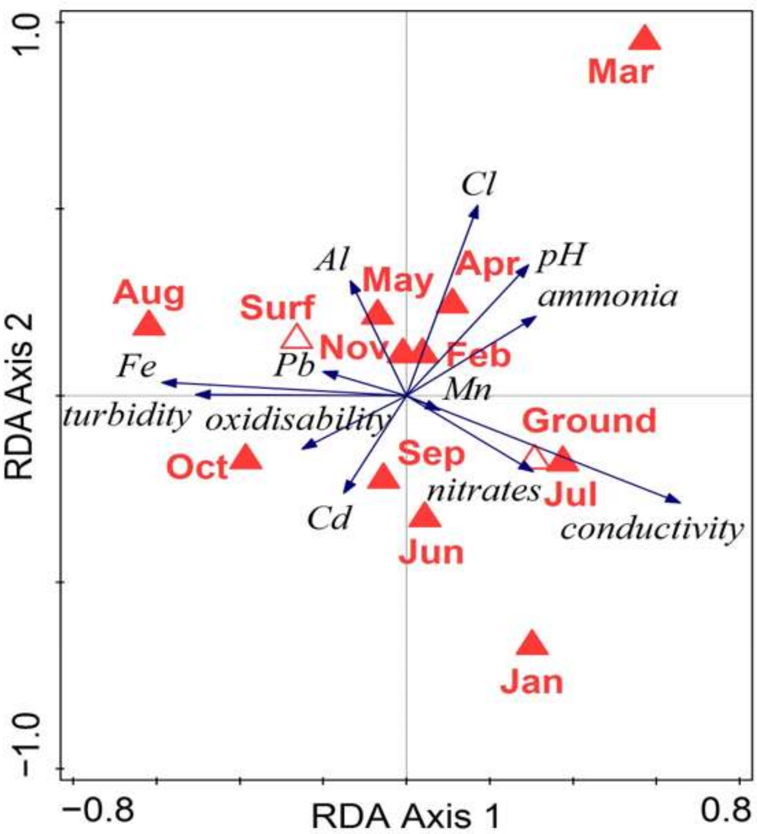


Figure 9. Graph of the redundancy analysis (RDA) of correlation of physicochemical parameters with water sources (surface and ground) and month of sampling
The month is abbreviated by the first three letters. The first two constrained axes are illustrated, but the second axis is not significant

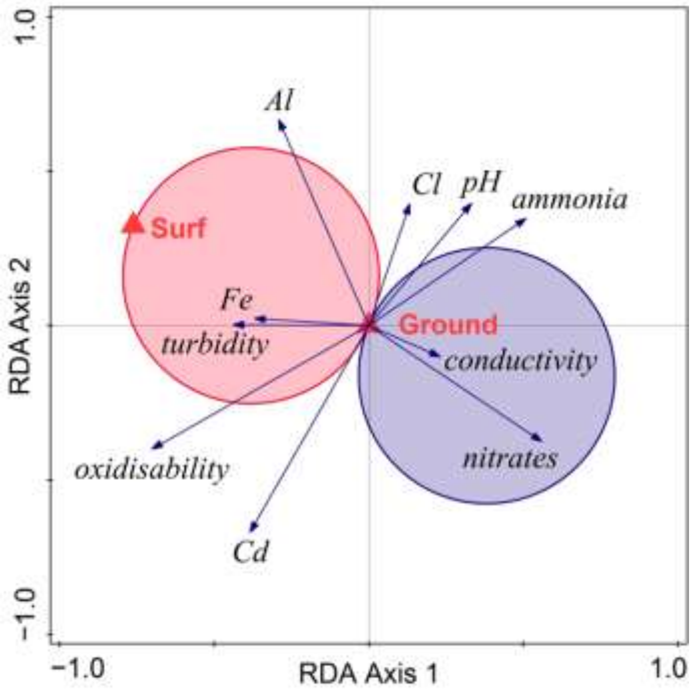


Figure 10. The graph with the t-value regarding the physicochemical parameters in relation to the water source (surface and groundwater). The pink circle delimits the ordering space for the significant positive response to the variable considered and the blue circle for the negative response.

The locality of origin of the drinking water samples explained 37.6% (23.2%) of the variation of the physicochemical parameters (pseudo-F = 2.6; p = 0.001), the first two constrained axes being significant, representing 39.6% (pseudo-F = 0.8, p = 0.001) and 27.1% (pseudo-F = 0.7, p = 0.002) of the explained variation (Figure 11). Along the first axis, conductivity and ammonia were negatively correlated with Fe and turbidity. Along the second axis, all response variables were positively correlated.

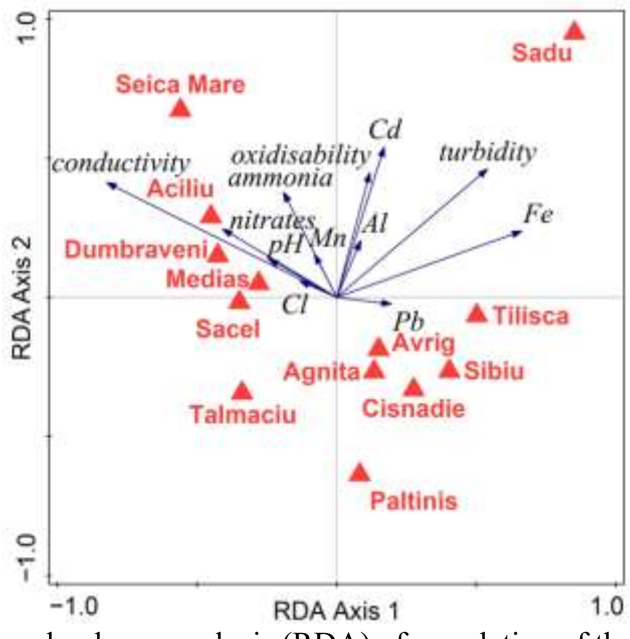


Figure 11. Graph of the redundancy analysis (RDA) of correlation of the physicochemical parameters with the locality (county of Sibiu). The first two constrained axes are illustrated, both being significant.

The co-inertia analysis between radioactivity and physicochemical parameters showed a negative correlation between global alpha activity and nitrates/oxidability/conductivity, and respectively a positive correlation with turbidity and residual chlorine. Global beta activity was positively correlated with conductivity, Cd, Mn and negatively with Fe (Figure 12).

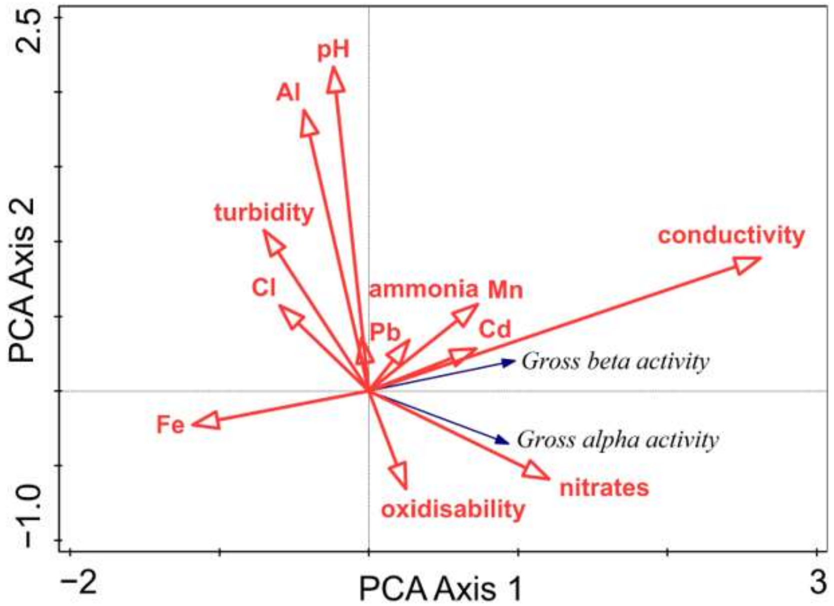


Figure 12. Graph of the co-inertia analysis between radioactivity (global alpha and global beta) and physicochemical parameters

4. Experimental research on physicochemical parameters and radioactivity indicators in drinking water from different sources in the county of Alba. Development of a multivariate model on drinking water quality.

Given the threat of nuclear accidents, conducting a complex analysis to understand how a space radioactivity parameter varies can be beneficial for identifying and mapping environmental risks that can affect multiple sources of drinking water. For example, geostatistical analysis of water quality data using advanced software is proposed as a useful tool in public health applications (PRO-WASH., 2022). Consequently, more data on the radiological characteristics of drinking water sources worldwide are needed.

The main objectives of this chapter of the doctoral thesis consisted in the evaluation of drinking water samples from different locations in the county of Alba from the point of view of physicochemical and radioactivity indicators, the complex statistical interpretation of the obtained results and the mathematical modelling, in order to create a spatial "profile" of the-temporal analysis of the radioactivity of drinking water in a county in Romania, as a useful tool for monitoring performance and establishing reference values, in the absence of a nuclear event.

4.1 Materials and methods

The analyses were carried out on a number of 65 drinking water samples from the county of SIBIU

- **Global alpha and beta radioactivity method:** according to SR ISO 9696/2018 and SR ISO 9697/2019 respectively;
- **Radon 222 concentration method:** according to SR EN ISO 13164-3:2020 standard;
- **Methods of physicochemical parameters:** according to the standards of each parameter;
- **Statistical analysis with the development of a mathematical model** (GLM, GLMM, RDA) using generalized linear model, generalized linear mixed models with gamma distribution and multivariate linear redundancy analysis.

4.2 Results and discussions

The data used for the present study were extracted from laboratory reports in the period 2017-2019 and 2022-2023, respectively 40 water samples analysed from the municipal distribution system (22.5%), springs (57.5%) and wells (20%).

Samples were collected annually from 7 locations illustrated in Figure 13, between January and November, as part of the National Drinking Water Quality Monitoring Program, meeting the eligible criteria for sampling points, locations, predetermined periodicity of sampling and accredited analytical methods.

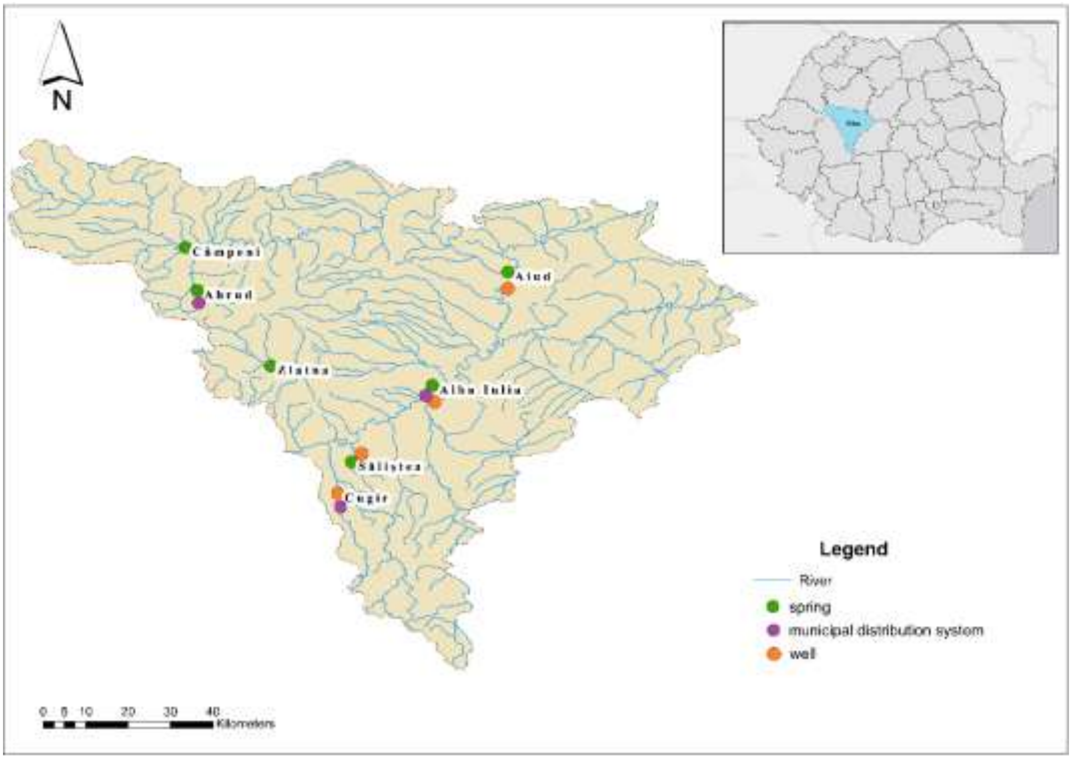


Figure 13. Location of sample collection points and water sources

4.2.1 Results regarding the radioactivity indicators of drinking water samples from different sources in the county of Alba

Table 6 highlights, in a comparative way with the study in the present doctoral thesis, the radioactivity parameters (average values) measured in different drinking water sources in several counties. Country-wide averages are also included.

Table 6. Parameters of drinking water radioactivity at local *vs.* national level (average values).

Area	**Source of water**	**Global Alpha Activity (Bq/L)**	**Global beta activity (Bq/L)**	**Rn-222 (Bq/L)**	**References**
Alba (Transylvania)	mixed	0.004	0.08	1.53	**This study**
Sibiu (Southern Transylvania)	mixed	0.01	0.09	4.1	(Tăban et al., 2023) – **another chapter of the doctoral thesis**
Galati (S-E region)	mixed	0.02	0.07	Not measured	(Pintilie et al., 2016)
Romania	mixed	0.025	0.12	2.23	(NIPH. Rep. 2022)

Compared to other regions or national levels in Romania, the county of Alba recorded lower values of Rn-222, global alpha and beta activity, despite the predominance of groundwater supply.

The highest values of Rn-222 in drinking water samples were recorded in Aiud, from two spring sources: 10.46 Bq/L in 2022 and 10.14 Bq/L in 2023, respectively.

4.2.2 Statistical analysis and development of a mathematical model (GLM, GLMM, RDA) on radioactivity indicators of investigated drinking water samples

The RDA analysis (Figure 14) showed that the localities from which the water samples were collected appeared to be the strongest predictors of radioactivity parameters. Through progressive selection, significant associations were identified for several localities, Aiud being the most influential (explains 28.84%, pseudo-F = 15.4, $p = 0.001$), indicating its substantial contribution to the model. Among the localities, Aiud had the highest levels of radioactivity, in contrast to the lower levels observed in the other localities in the county of Alba.

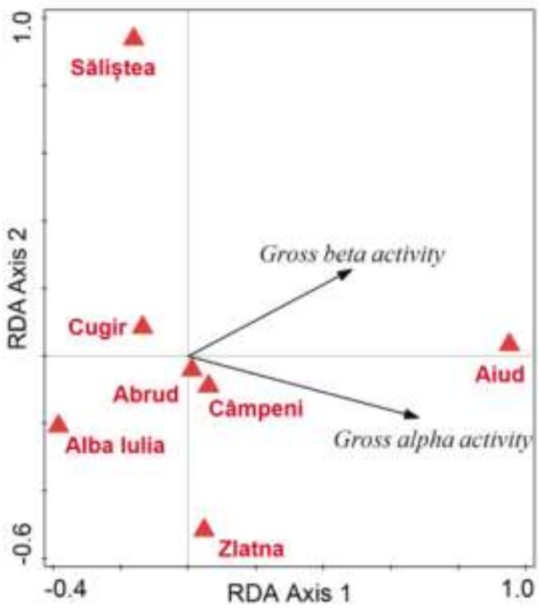


Figure 14. Redundancy analysis graph (RDA) regarding the correlation between global alpha/beta radioactivity values and locality (county of Alba).

Analyzing the influence of localities only on global alpha radioactivity using the GLM model with gamma distribution (Figure 15), we tested the difference from the reference value, i.e. the locality that recorded the lowest global alpha radioactivity, namely Săliștea. Almost all the analyzed localities (except Alba Iulia) showed significantly higher values of global alpha radioactivity (Abrud: $t = 3.443$, $df = 22$, $p < 0.001$; Aiud: $t = 7.059$, $df = 22$, $p < 0.001$; Câmpeni: $t = 4.027$, $df = 22$, $p < 0.005$; Cugir: $t = 2.223$, $df = 22$, $p < 0.05$; Zlatna: $t = 4.278$, $df = 22$, $p < 0.001$).

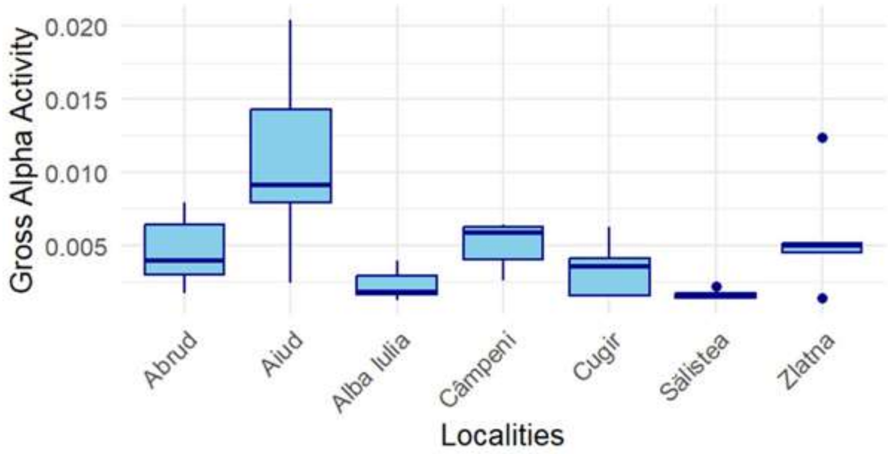


Figure 15. Diagram regarding the global alpha radioactivity in the localities of the county of Alba taken in the study

4.2.3 Correlation analysis of the radioactivity of drinking water samples with the physicochemical parameters investigated.

Based on the Spearman correlation analysis (Figure 16), moderate to statistically strong correlations were identified between certain pairs of variables. Thus, there is a statistically significant positive correlation between global alpha and beta radioactivity ($p = 0.0075$, $r = 0.42$), as well as between nitrates and ammonia ($p = 0.0001$, $r = 0.57$), nitrates and total hardness ($p = 0.0002$, $r = 0.56$), nitrates and nitrites ($p = 0.001$, $r = 0.50$), nitrites and total hardness ($p = 0.0063$, $r = 0.42$), ammonia and total hardness ($p = 0.0133$, $r = 0.39$). Moreover, there is a statistically significant correlation between global beta radioactivity and Rn-222 ($p = 0.0018$, $r = 0.72$). In addition, weaker correlations are observed between other variables, such as turbidity and global alpha radioactivity ($p = 0.0110$, $r = 0.40$) or global beta radioactivity ($p = 0.0419$, $r = 0.32$), as well as between Rn-222 and turbidity ($p = 0.0918$, $r = 0.44$).

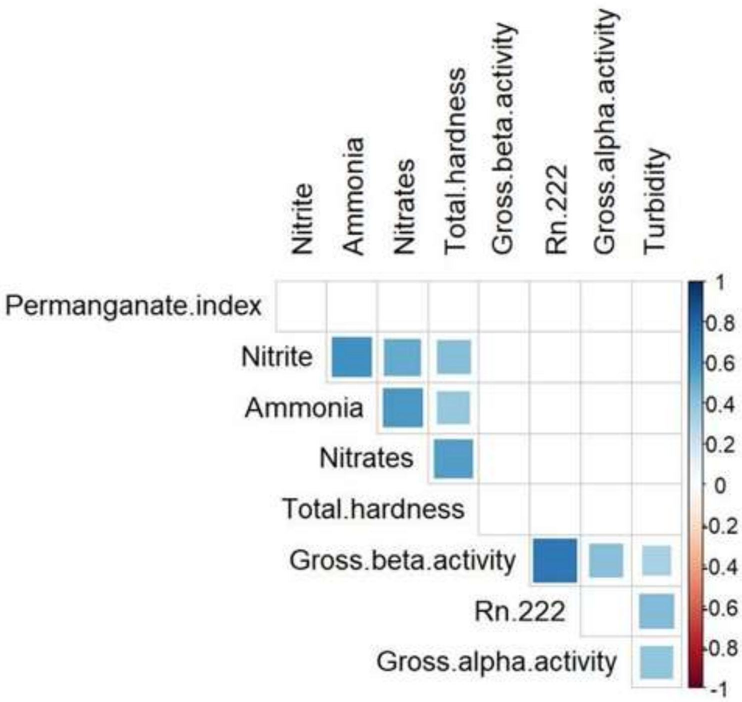


Figure 16. Correlation coefficients between radioactivity and physicochemical parameters of drinking water samples from the county of Alba.

The color blue represents a positive correlation, with darker shades indicating stronger correlations (closer to 1). Larger squares correspond to lower p-values (< 0.05). Only significant (p < 0.05) or marginally significant (0.05 < p < 0.1) results are shown on the graph.

As illustrated in Figure 17, the RDA redundancy analysis revealed very statistically significant relationships, supported by the Monte Carlo permutation test for both the first canonical axis (F ratio = 1.1, $p = 0.001$) and all canonical axes (F ratio = 3.1, $p = 0.001$). Significant effects were observed for the source, as follows: wells (explains 12.9%, pseudo-F = 5.6, $p = 0.004$), municipal distribution system (explains 10.4%, pseudo-F = 4.4, $p = 0.004$), spring (explains 5.1%, pseudo-F = 2.0, $p = 0.06$); localities: Abrud (explains 8.8%, pseudo-F = 3.7, $p = 0.03$) and Aiud (explains 7.6%, pseudo-F = 3.1, $p = 0.05$).

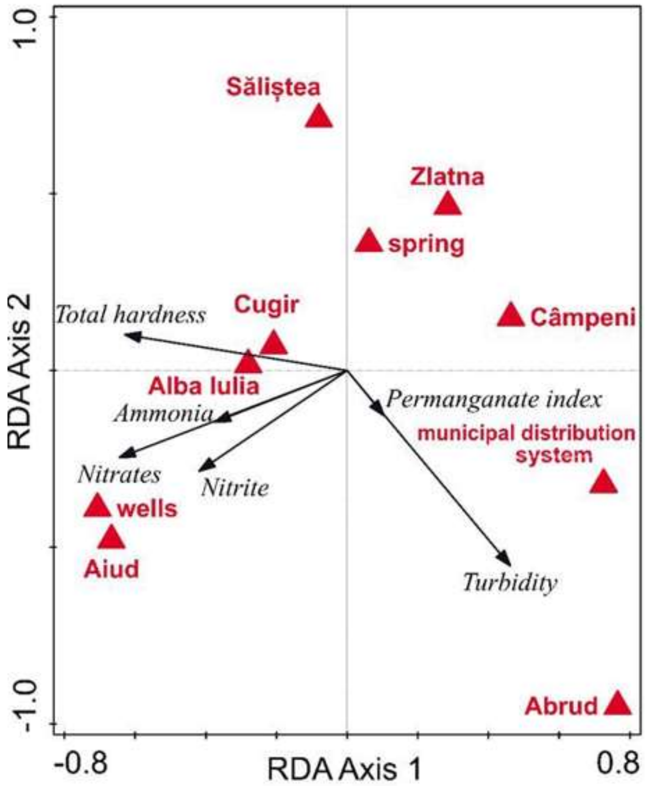
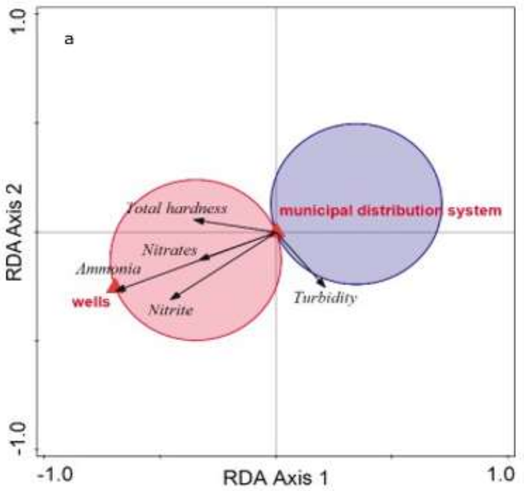


Figure 17. RDA graph on the relationship between physicochemical parameters and the type of water sources and locality.

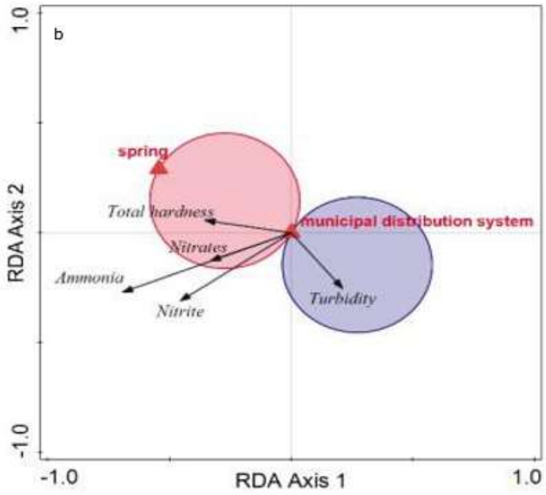
Significant variations in water quality parameters were found for different areas. Thus, the localities of Aiud, Alba Iulia and Cugir had the highest levels of total hardness, ammonia, nitrites and nitrates, while Campeni and Abrud had higher values of the permanganate index and high levels of turbidity.

Using van Dobben circles to compare water sources, the following results emerged:

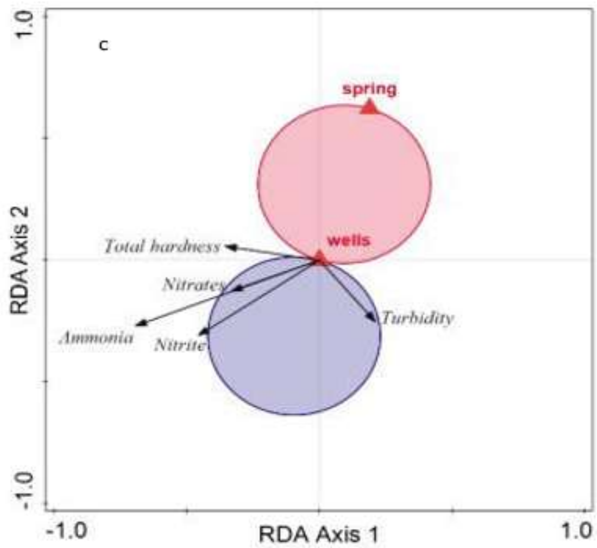
1. The concentrations of ammonia, nitrites, nitrates and total hardness are higher in the wells compared to the values for samples from the municipal distribution system (Figure 18 a);
2. Total hardness and nitrates have higher values, and turbidity shows a negative response when comparing the springs with the municipal distribution system (Figure 18 b);
3. Turbidity and nitrates show a negative response when comparing springs to wells (Figure 18 c).



(a)



(b)



(c)

Figure 18. Comparisons of physicochemical parameters: a) wells compared to the municipal distribution system, b) springs compared to the municipal distribution system and c) springs compared to wells. Within the biplots, the pink circle delimits the ordering space for significant positive responses to the variables considered, while the blue circle represents the negative response.

Of the 40 drinking water samples from the county of Alba investigated in the present chapter of the doctoral thesis, 9 samples (22.5%) exceeded the allowed values for nitrates, the water from the wells being the main contributor.

5. Comparative study on global alpha/beta radioactivity and the effective radiation dose from drinking water ingestion in the counties of Sibiu and Alba

5.1 Evaluation of the effective radiation dose attributed to the global alpha and beta radioactivity measured in drinking water samples in the counties of Sibiu and Alba, within the period 2000-2016

The effective dose, not the total one, but assumed to the contribution of each radionuclide was calculated according to the formula (Gorur and Camgoz, 2014; Pintilie et al., 2016a):

$$D_{ef} = \Lambda_x \times R \times CF \text{ (Sv/year)}$$

Where:

Λ_x = alpha and beta-global radioactivity, corresponding to the type of radionuclide assumed, (Bq/L);

R = annual drinking water intake rate for adults, 730 (L) (WHO, 2017);

CF = dose-specific activity conversion factor, corresponding to the assigned isotope, (Sv/Bq)

$C_{(assum.\ Po\text{-}210)}$ – the concentration assumed as a specific activity of ^{210} Po, as coming from the entire determined global alpha activity. (1.2×10^{-6}) (IAEA, 2014);

$C_{(assum.\ Ra\text{-}226)}$ – the concentration assumed as specific activity of ^{226} Ra, as coming from the entire determined global alpha activity (2.8×10^{-7}) (IAEA, 2014, Table III 2D);

$C_{(assum.\ Pb\text{-}210\ and\ Ra\text{-}228)}$ – the concentration assumed as specific activity of ^{228}Ra and ^{210}Pb, respectively, as coming from the entire determined global beta activity (6.9×10^{-7}) (IAEA, 2014).

This relationship uses assumed concentrations of alpha-emitting radionuclides, as originating from determined global alpha activity, and assumed concentrations of beta-emitting radionuclides, as originating from determined global beta activity.

Figures 19-20 show the values for the average effective dose attributed to global alpha and beta radioactivity, calculated for the period 2000-2016, for both Sibiu and Alba counties.

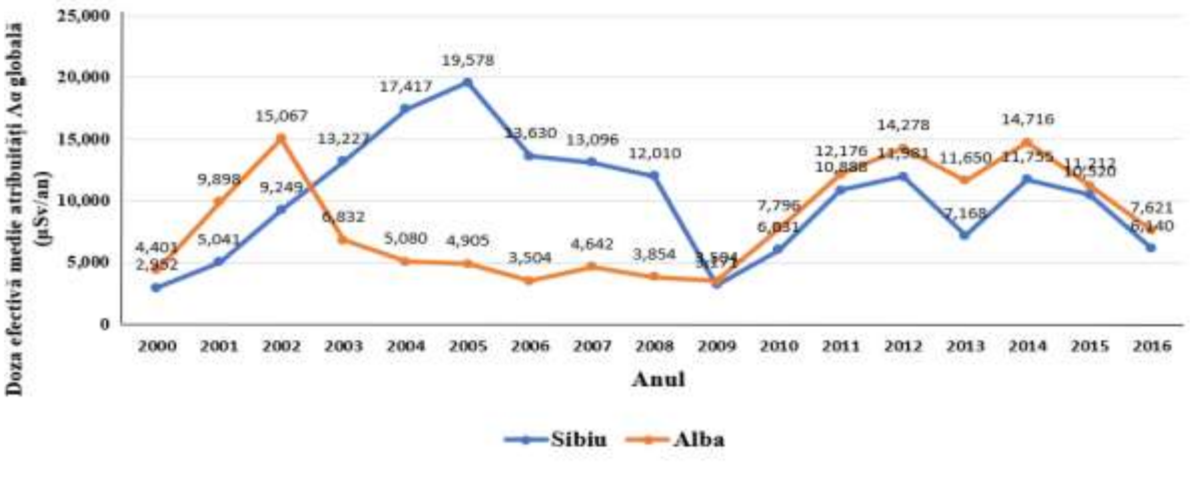


Figure 19. Average effective dose attributed to global alpha radioactivities of water samples taken from the counties of Sibiu and Alba in the period 2000-2016

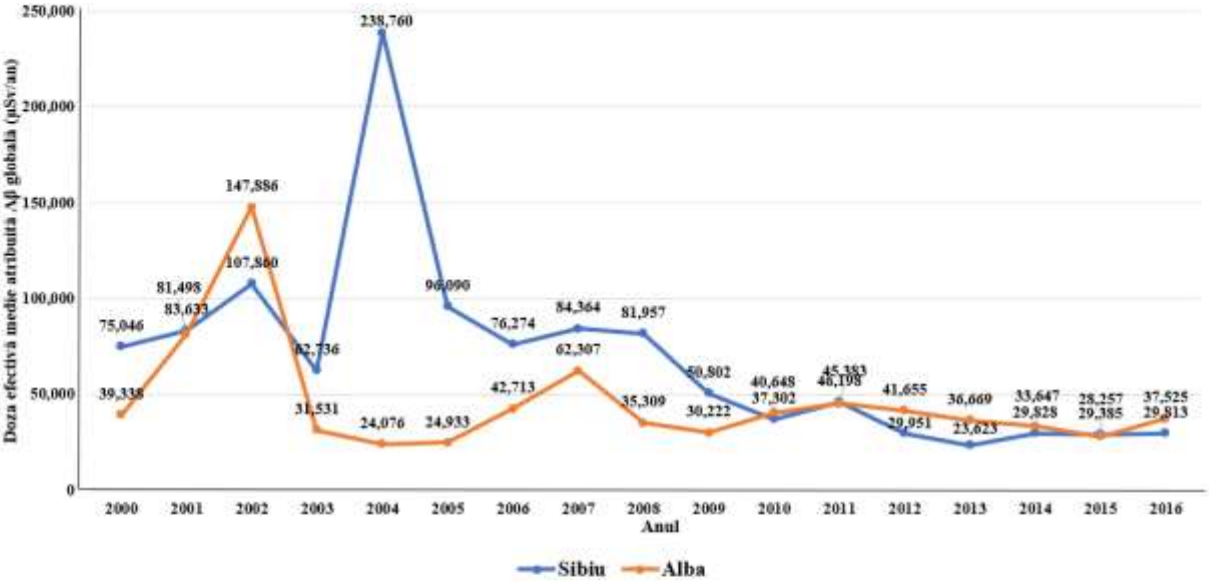


Figure 20. Average effective dose attributed to global beta radioactivity of water samples taken from the counties of Sibiu and Alba between 2000 and 2016.

In the study on the radioactivity of drinking water samples in the county of Sibiu, the calculated average values of the effective annual dose attributed to the global beta radioactivity from the radiometric measurements for the period 2000-2016, are between 23.62 µSv/year and 238.76 µSv/year, using the concentration assumed as specific activity of ^{228}Ra, respectively ^{210}Pb, as coming from the entire determined global beta activity (6.9 x 10^{-7}). This high average value of 238.76 ± 28.112 µSv/year in 2004 is due to the fact we have noted an increased value for a drinking water sample (Apoldu de Sus) above the recommended limit (Law 301/2015) of 1 Bq/L for global beta activity. The mean dose values using the concentration assumed as specific activity of ^{210}Po, as coming from the entire determined global alpha activity (1.2 x 10^{-6}) ranged from 2.95 µSv/year to 19.57 µSv/year.

In the study on the radioactivity of drinking water samples in the county of Alba, during the period 2000-2016, the average values of the effective annual dose attributed to the global beta activity, ranged from 24.07 µSv/year to 147.88 µSv/year, using the concentration assumed as specific activity of ^{228}Ra, respectively ^{210}Pb, as coming from the entire determined global beta activity (6.9 x 10^{-7}). This value of 147.88 ± 21.910 µSv/year in 2002 is due to the increased value close to the recommended limit of 1 Bq/L of the global beta activity in the drinking water sample from Sântimbru. The mean dose values using the concentration assumed as specific activity of ^{210}Po, as coming from the entire determined global alpha activity (1.2 x 10^{-6}) ranged from 3.50 µSv/year to 15.06 µSv/year.

For the radiometric effective dose assessment method applied in this study, only global alpha and beta activity were used, as they show rapid response time. In the case of this study, the contribution of ^{40}K, which is attributed to beta-emitting radionuclides, is not subtracted.

5.2 Comparative study of the average values of global alpha and beta radioactivity of drinking water samples collected from the counties of Sibiu and Alba

The average values of global alpha and beta radioactivity, over the investigated period of 17 years (2000-2016) are presented comparatively in Figures 21 and 22.

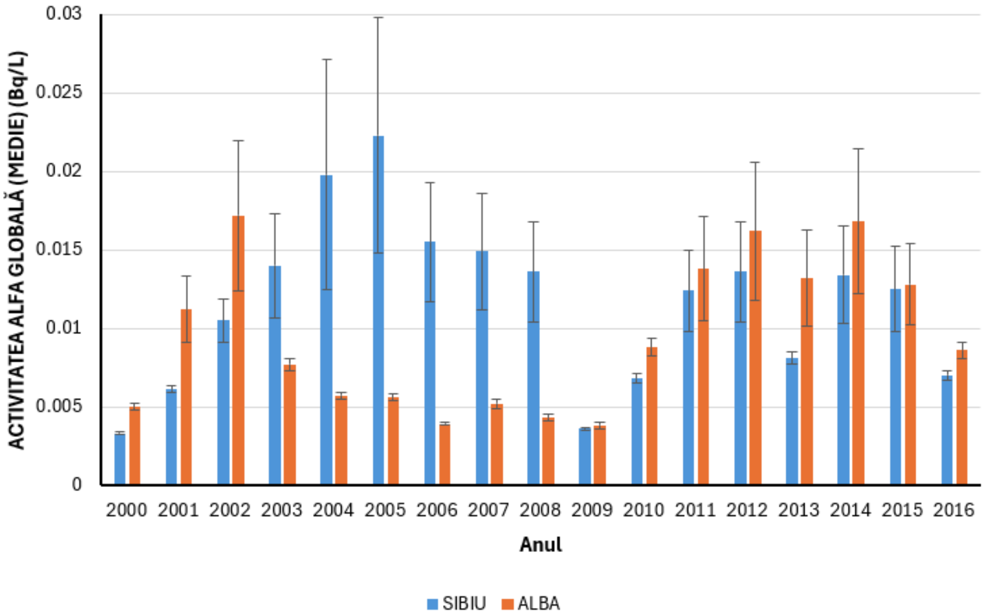


Figure 21. Comparative presentation of the average values of global alpha radioactivity in drinking water samples, period 2000-2016, for the counties of Sibiu and Alba.

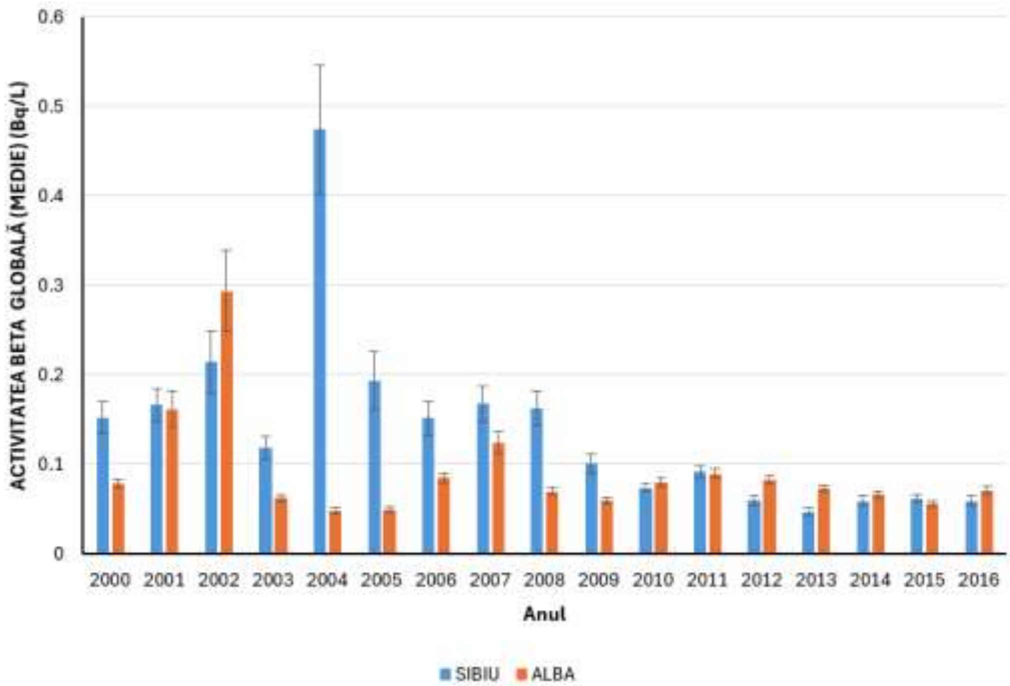


Figure 22. Comparative presentation of the average values of global beta radioactivity in drinking water samples, period 2000-2016, for the counties of Sibiu and Alba.

For the county of Sibiu, the highest average value of global alpha radioactivity was 0.0223±0.0075 Bq/L in 2005, and the lowest average value was 0.0033±0.0001Bq/L in 2000. As for the global beta radioactivity, the highest average value was 0.4740±0.0072 Bq/L in 2004, and the lowest average value was 0.0468±0.0039 Bq/L recorded in 2013.

For the county of Alba, the highest average value of global alpha activity was 0.0172±0.0048 Bq/L in 2002, and the lowest average value was 0.0038±0.0002 Bq/L in 2009. Regarding global beta radioactivity, the highest average value was 0.2935±0.00454 Bq/L, and the low average value was 0.0477±0.0033 Bq/L.

The average values for global alpha and beta radioactivity of drinking water monitored between 2000 and 2016 in the two neighbouring counties are within the limit provided by the national law (Law no. 301/2015), respectively 0.1 Bq/L for global alpha activity and 1 Bq/L for global beta activity, and by WHO (WHO, 2011), 0.5 Bq/L for global alpha activity and 1 Bq/L for global beta activity.

Figures 23 and 24 illustrate the global beta radioactivity, in which the values close to and those exceeding the maximum limit provided by the national law (Law no. 301/2015 and Law no. 458/2002), the European law (Directive 51/2013) and the WHO (WHO, 2017) are presented, both for the county of Sibiu and Alba.

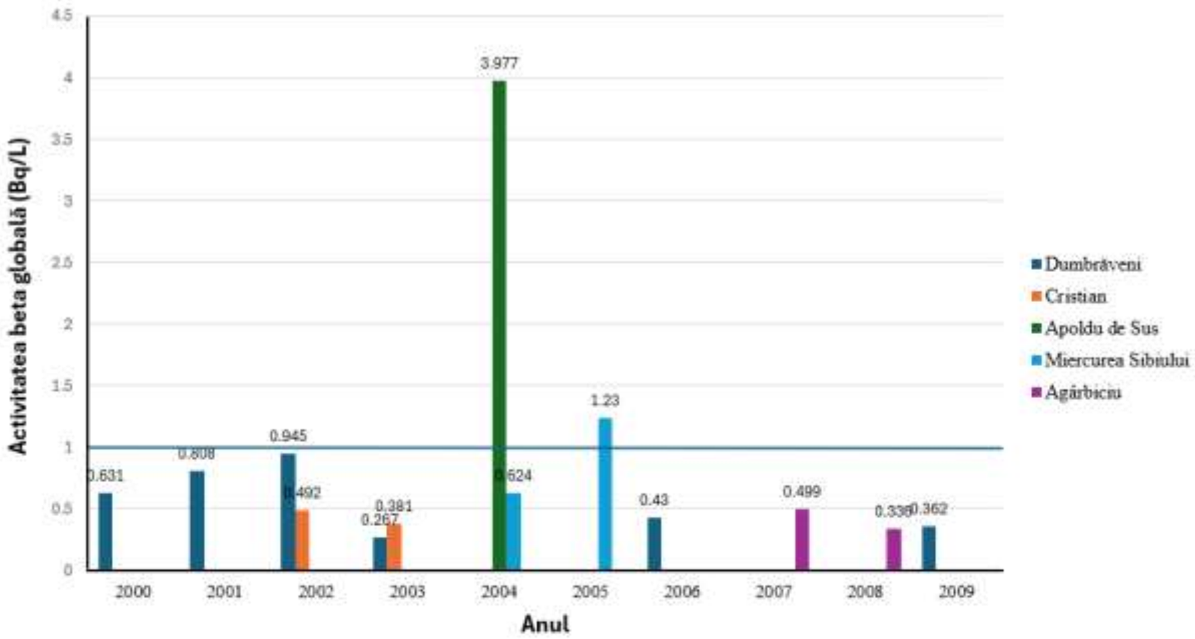


Figure 23. Global beta radioactivity of drinking water samples from the county of Sibiu, which recorded values close to, and respectively values that exceeded or are at the limit of the maximum allowed concentration.

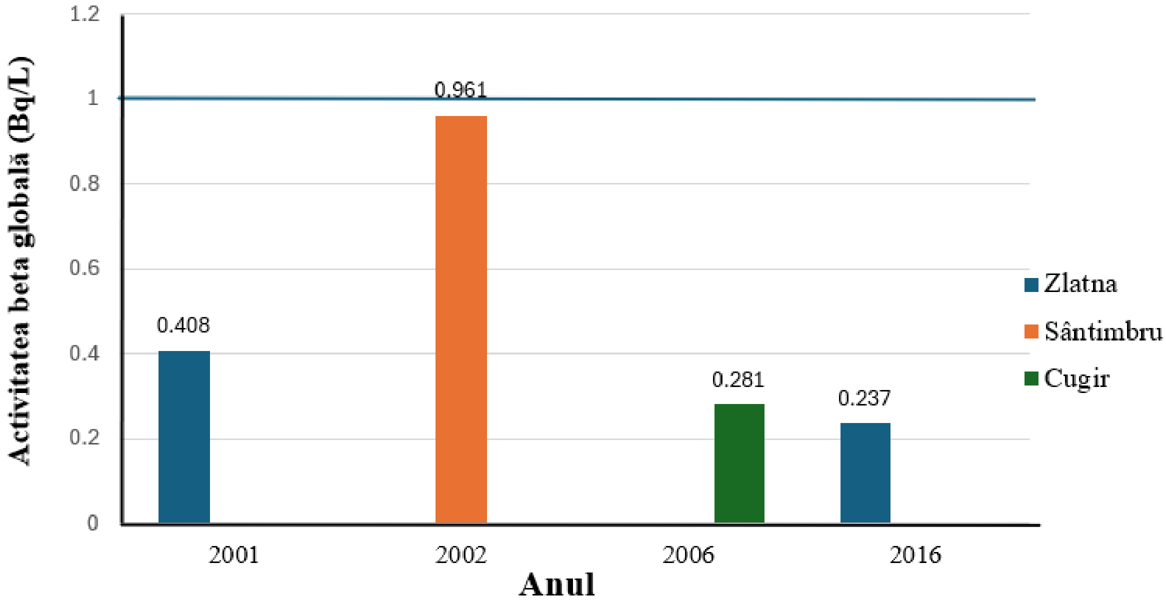


Figure 24. Global beta radioactivity of drinking water samples from the county of Alba, which recorded values close to, and respectively values that exceeded or are at the limit of the maximum allowed concentration.

In the county of Sibiu, in 2004, increased values in the case of global beta radioactivity of 3.9770±0.8990 Bq/L in the Apoldu de Sus drinking water sample were recorded, above the maximum limit provided by the national law and the WHO. In the county of Alba, the relatively high average value in 2002 is due to the increased value of global beta-activity of 0.9610±0.0896 Bq/L in the drinking water sample from Sântimbru, being very close to the maximum limit provided by the WHO (WHO, 2011), 0.5 Bq/L for global alpha activity and 1 Bq/L for global beta activity.

Concluding remarks

- From the descriptive research on the quality of drinking water distributed in a centralized system at national level for the period 2017-2021, it can be concluded that exceedances of the values of some physicochemical parameters (ammonium, nitrates, manganese, iron, pH, turbidity) were recorded for water from public wells and springs in certain counties all over the country.

- The results obtained from the experimental study for the evaluation of beta-emitting radioisotopes in the drinking water from the counties of Sibiu and Alba showed that the level of radionuclides ^{137}Cs and ^{90}Sr in water samples from the county of Sibiu, in the period 2000-2003, indicates lower average values than those in drinking water samples from the county of Alba.

- The results obtained from the experimental study for the evaluation of alpha-emitting radioisotopes in the drinking water from the counties of Sibiu and Alba showed that the level of radionuclide ^{226}Ra in water samples from the county of Sibiu, in the period 2000-2005, indicates higher values than in drinking water samples from the county of Alba.

- Results on the assessment of global alpha activity in drinking water (65 determinations); the highest global alpha activity was recorded in the drinking water collected from Tilișca (surface water) while the lowest value of global alpha activity was recorded in the drinking water in the municipality of Sibiu (surface water).

- Results regarding the global beta activity in drinking water over a period of 5 years (65 determinations) point out the highest global beta activity in the drinking water from Tilișca in September 2017 and the lowest value in the drinking water from Păltiniș (water from the captured springs) in June 2018.

- The average value of the concentration of ^{222}Rn from 13 drinking water samples collected over a period of 2 years (26 determinations) highlights the lowest concentration of ^{222}Rn in the surface drinking water of the town of Avrig, while the highest values were found in the drinking water samples of Păltiniș (deep water), due to the fact that it is groundwater; these values exceeded the US-EPA reference value of 11.1 Bq/L.

- The study allowed the development of a multivariate spatio-temporal model of a number of 65 drinking water samples from 13 supply areas located in the county of Sibiu; the elaborated model, shows correlations and variability of radiological and physicochemical quality parameters in relation to the water source, location and collection period, and indicates the water source as the best predictor in the GLM model with gamma distribution.

- The results of the RDA analysis on the radiological parameters of the water samples investigated from the 13 locations of the county of Sibiu indicate that the radioactivity of the water increased from surface to deep and ground sources.

- The statistical analysis showed a significant positive correlation of alpha activity with global beta activity, both positively correlated with conductivity, global alpha activity positively correlated with nitrates and negatively correlated with pH, while global beta activity is positively correlated with manganese and negatively correlated with iron, these correlations being also confirmed by co-inertia analysis; the results are significant for turbidity and iron in surface waters, for conductivity and nitrates in groundwater, with a special interest for the rural localities of Sadu and Şeica-Mare.

- The increased levels of some chemical contaminants, such as ammonium and iron, in the water samples investigated may be of concern, especially for ammonium, in rural localities due to agricultural practices and pastoral conditions in the area.

- The results regarding the assessment of the global alpha activity in drinking water (40 determinations) over 5 years in the county of Alba, indicated the highest global alpha activity for drinking water collected from Aiud in 2022 and the lowest value of global alpha activity in the drinking water in the Alba-Centre area in 2019.

- The results on the global beta activity in drinking water over a period of 5 years (40 determinations) in the county of Alba, indicated the highest global beta activity for drinking water in Aiud in 2023 and the lowest value in drinking water in Zlatna in 2023.

- The average value of the concentration of ^{222}Rn of the 8 drinking water samples over a period of 2 years (16 determinations) highlights the lowest concentration of ^{222}Rn in the drinking water of Cugir and the highest value in the drinking water of Aiud (2022), close to the US-EPA recommended reference value of 11.1 Bq/L.

- The RDA analysis on the radiological parameters in the water samples investigated from the 8 localities of the county of Alba revealed significantly higher values of global alpha activity. Among the localities, Aiud had the highest levels of radioactivity, in contrast to the lower levels observed in other localities.

- Significant variations in the physicochemical parameters of water quality were obtained in the localities of Aiud, Alba Iulia and Cugir, which presented the highest levels of total hardness, ammonia, nitrites and nitrates, while the localities of Câmpeni and Abrud presented higher values of the permanganate and turbidity index.

- The results of the effective annual dose attributed to the radioactivity of water show that both in the county of Sibiu and Alba there were higher average values of the dose attributed to global beta activity compared to the values attributed to global alpha activity. In the case of this study, the contribution of 40K, which is attributed to beta-emitting radionuclides, was not subtracted.

- In the county of Sibiu, the average increase in 2004 of global beta activity is due to the high value of global beta radioactivity of the drinking water sample from Apoldu de Sus, which exceeded the WHO recommended limit of 1 Bq/L.

- In the county of Alba, the average value of the global beta activity increased in 2002 is due to the high value of the global beta-activity in the drinking water sample from Sântimbru, being very close to the maximum limit of 1 Bq/L.

Original contributions

Based on the methodology used, measurements, and the encouraging results obtained, the present study is based on personal contributions, both theoretical and practical, materialized through publications such as articles, participation in international conferences, offering a perspective on some potential risks to which the population is exposed by the consumption of inadequate water from a radiological or physicochemical point of view. Another aspect related to the importance of this study is to raise awareness of the importance of continuous and rigorous monitoring of the essential parameters of drinking water quality.

- ✓ *Investigation of the content of the radioactive elements Cs-137, Sr-90 and Ra-226 in the drinking water from the counties of Sibiu and Alba, during the period 2000-2003;*

- ✓ *Investigation of the physicochemical parameters and radioactivity indicators of a number of 65 drinking water samples, over a period of 5 years, 2017-2021 from the county of Sibiu, and respectively of a number of 40 drinking water samples from the county of Alba, over a period of 5 years, 2017-2019 and 2022-2023;*

- ✓ *Statistical analysis and development of a multivariate spatio-temporal mathematical model regarding the physicochemical and radioactivity indicators of drinking water (GLM, GLMM, RDA);*

- ✓ *Assessment of the effective radiation dose in population attributed to the global alpha and beta radioactivity of drinking water samples from the counties of Sibiu and Alba for a significant period of time (2000-2016);*

- ✓ *Comparative study of the average values of global alpha and beta radioactivity, for a period of 17 years, (2000-2016) in water samples from the counties of Sibiu and Alba.*

Future research directions

This study opens the following research directions:

- ✓ *Study of the levels of artificial radionuclides in drinking water and the assessment of the corresponding effective annual dose, as well as the evaluation of the detriment to health caused by their ingestion;*

- ✓ *Research on the correlation between the radioactive content of deep water and the geological composition of the soils from which it originates;*

- ✓ *Assessment of the population's exposure to ionizing radiation from other sources (e.g., building materials, medical exposure);*

- ✓ *Improvement of the radiochemical separation method to determine the radionuclide content in water, namely by shortening the determination time;*

- ✓ *Assessment of the concentration of ^{40}K in water samples, which plays an important role in global beta activity.*

Keywords: global alpha/beta radioactivity, physicochemical indicators, ^{222}Rn concentration, radioactive elements ^{137}Cs, ^{90}Sr and ^{226}Ra, maximum allowable concentration (MAC).

List of publications resulting from doctoral research, published or accepted for publication

- **Articles published in ISI listed journals**

1. **Tăban C.I.,** Benedek A.M., Stoia M., Cocîrlea M.D., Oancea S., A Multivariate Model of Drinking Water Quality Based on Regular Monitoring of Radioactivity and Chemical Composition, *Applied Sciences* Basel, 13, (18), 10544, DOI10.3390/app131810544, (2023).https://www.mdpi.com/2076-3417/13/18/10544

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2. **Tăban C.I.,** Sandu A., Oancea S., Stoia M., Gross alpha/beta radioactivity of drinking water and relationships with quality parameters of water from Alba county, Romania, *Romanian Jorunal of Physics*, vol. 69, nr. 7-8, (2024). https://doi.org./10.59277/RomJPhys.2024.69.806.

- **Articles published in Proceedings ISI**

1. **Tăban C.I.,** Tăuşan I., Oancea S., Radiochemical Analysis, Gross Alpha and Gross Beta Radioactivity, ^{137}Cs and ^{90}Sr Concentration of Milk Samples from SIBIU and ALBA County, ROMANIA, *Proceedings of 22nd International Multidisciplinary Scientific Geo Conferince SGEM 2022*, Publisher STEF92 Technology, 22(5.1), 10.5593/sgem2022/5.1/s20.043, (2022). https://epslibrary.at/sgem_jresearch_publication_view.php?page=view&editid1=8707

- **Papers published in BDI indexed journals**

1. **Tăban C.I.,** Oancea S., Research on radon (^{222}Rn) content in drinking-water samples collected from Sibiu County, *Current Trends in Natural Sciences,* 10(20), 212-218, (2021) https://doi.org/10.47068/ctns.2021.v10i20.029 ISSN: 2284-953X, ISSN-L: 2284-9521.

2. **Tăban C.I.,** Moșteanu A.M., Oancea S., Study on Gross Alpha and Beta Radioactivity of Samples of Bottled Waters in Romania, *Current Trends in Natural Sciences*, 13(25), (2024).

- **Papers presented at international conferences**

1. **Tăban C.I.**, Cirican T.M., Oancea S., Contribution Regarding the Radioactive Contamination of Drinking Water: Health Concern, Regulations, Methods of Assessment, *The XVI-th International Student Symposium IF-IM-CAD*, lucrare publicată în revista, *Journal of Young Scientist*, Volume IX, pg. 59-68, (2022). *https://journalofyoungscientist.usamv.ro/index.php/scientific-papers/622-contribution-regarding-the-radioactive-contamination-of-drinking-water-health-concern-regulations-methods-of-assessment*
2. **Tăban C.I.,** Oancea S., Research on radon (^{222}Rn) content in drinking-water samples collected from Sibiu County, *The International Scientific Symposium Current Trends in Natural Sciences* 28-30 mai 2021, University of Science and Technology POLITEHNICA Bucharest, Pitești University Centre, Romania. https://www.natsci.upit.ro/international-symposium/ (paper published in the BDI indexed journal).

3. **Tăban C.I.**, Moșteanu A.M., Oancea S., Study on Gross Alpha and Beta Radioactivity of Samples of Bottled Waters in Romania, *The International Scientific Symposium Current Trends in Natural Sciences* 16-18 mai 2024, University of Science and Technology POLITEHNICA Bucharest, Pitești University Centre, Romania. https://www.natsci.upit.ro/international-symposium/ (paper published in the BDI indexed journal).

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