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THESIS ABSTRACT

CONTRIBUTIONS TO STUDY OF AMBIENT ENVIRONMENTAL RADIOACTIVITY

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Thanks

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CONTENTS

PART I - DOCUMENTARY STUDY (LITERATURE REVIEW)

PART II -A EXPERIMENTAL STUDY AND ORIGINAL CONTRIBUTIONS

(numbering of tables, figures and references are made according to thesis)

ABSTRACT

More than a century after the discovery of natural radioactivity by Henri Becquerel in 1896, purification and identification of radium and polonium by the Curies in 1898 and the discovery by the spouses Joliot-Curie of artificial radioactivity in 1934, radioactivity represents an vanguard domain, covering a wide spectrum of civil and military applications.

Development of nuclear energy and radioprotection by studying the elements radiotoxicity have provided solid scientific basis for regulating the nuclear fuel cycle operations and emissions of radionuclides in the environment [1].

Radiation from ionizing radiation sources have become a necessary part of everyday life, covering areas such as food, medicine, biology, agriculture, etc. [2].

Also, natural radioactivity (cosmic radiation, terrestrial radioactivity, radon) and artificial (derived from nuclear weapons tests, nuclear accidents, the activity of nuclear power plants and other nuclear units) currently existing in nature creates a complex environment for living organisms, including humans [3], [4].

In view of the fact that radioctivity both natural and artificial can be seen in all enviromnetal compartments, the thesis addresses a wide range of analysis (total beta, beta and gamma spectrometry and gamma dose rate) and different type of samples: air (by gamma dose rate, atmospheric aerosols, total atmospheric deposition and precipitation), water, soil and spontaneous vegetation. Thesis was developed through the need to solve practical problems and work efficiency and also interpretation of data on the environmental radioactivity monitoring.

The main objective of this work was to study the dynamics of environmental radioactivity. This has been developed and pursued through several secondary objectives such as: analysis between the environmental radioactivity and meteorological variables at different time intervals, estimation and prediction of radionuclide concentrations in different time series analyzed using multiple linear regression model, adaptation and optimization of the strategy for environmental radioactivity monitoring.

The thesis is structured into two sections. **The first section**, divided into four chapters, contains a schematic overview of the information found in the literature relevant to the topic of study chosen and **the second section**, divided into seven chapters, contains original results.

Chapter 1 contains theoretical notions of radiation and their ways of entering the environment. Also are presented, according to their origin, provenance and content, the average values of natural radioactivity from air, land, water, sediment, food and cosmic radiation and the average values of artificial radioactivity, mainly due to testing of nuclear weapons during the 1940's–1960's and due to the operating nuclear power plants.

Chapter 2 presents the radioactive isotopes circuit in the environment, pollution sources and major radionuclides pathways different media to humans. Also, are shown radionuclide transport through the environment, based on the ecosystem passed (land, water and air) and the radionuclide's concentrations and transfer factors.

Chapter 3 presents the main technical methods for detecting and measuring radioactive elements activity, classifying detectors based on physical principles underlying constructive radiation measurement. In this chapter is made also an overview of the different methods used for detection and measurement of ^{222}Rn, ^{220}Rn, ^{3}H, ^{137}Cs and ^{131}I.

Chapter 4 deals with the problem regardind the effects over matter's and humans exposure to radiationThere are presented the effects of radiation when the source is outside or inside of the organism. Also, there is shown the metabolism of radionuclides to living organisms in general and to humans in particular, in terms of toxicity radionuclides.

Chapter 5 is dedicated to the **study on optimisation of water samples preparation in order to determine the tritium volumic activity**. This chapter presents an alternative method to the classical one, found in the literature (ISO 9698:2010 - "Water quality - Determination of tritium activity concentration - Liquid scintillation counting method" [90]), for the preparation of water to determine the tritium volumic activity. In order to demonstrate that the proposed method (unstandardized method) optimises the sample preparation stage, keeping the same level of confidence of the results, test results were done in parallel for the two methods, checking for each the efficiency of method criteria (instrumental detection limit, precision, accuracy, recovery rate and robustness).

Sampling

For the purposes of this study, there were used synthetic samples obtained from bidistilled water spiked with T in the form of HTO.

Homogeneity and stability testing for spiked sample were performed in accordance with Annex B of ISO 13528:2005 [91]. The results of the tests of homogeneity and stability were within the criteria required by this standard in both, container and between containers.

Materials and Methods - standardized method

Reagents

- Sodium carbonate (Na_2CO_3), anhydrous
- Sodium thiosulfate ($Na_2S_2O_3$), anhydrous
- Reference water with tritium volume activity below 0.5 Bq / L
- Scintillation solution was used scintillation cocktail Ultima Gold LLT tritium with the determining efficiency of about 30% and a very low level of background.
- Certified reference material - open source standard for tritium
- Bidistilled water

Apparatus

- For sample preparation: Analytical balance, heating mantles, with temperature control and magnetic stirrer; oven with temperature control, bidistilled water system;
- For sample analysis: analyzer with ultra low background liquid scintillation;

Preparation of samples

In a round-bottomed flask are placed 250 mg of $Na_2S_2O_3$, 500 mg of Na_2CO_3 and 250 mL of sample. For spiked samples, they have a theoretical activity of 2.8070 Bq for 250 mL. The system installation was made and the distillation was conducted slowly with increasing temperature, resulting small residues. First part of the distillate was discarded, the middle part of the distillate was colected for analisys and the last part of the distillate and the residue was discarded. 10 ml of the distillate obtained was put into a counting vial, then added 10 mL of scintillation cocktail.

Materials and Methods - unstandardized method

Reagents

- Sodium carbonate (Na_2CO_3), anhydrous
- Sodium thiosulfate ($Na_2S_2O_3$), anhydrous
- Reference water with tritium volume activity below 0.5 Bq / L
- Scintillation solution was used scintillation cocktail Ultima Gold LLT tritium with the determining efficiency of about 30% and a very low level of background.
- Certified reference material - open source standard for tritium
- Bidistilled water

Apparatus

- For sample preparation: Analytical balance, rotary evaporation; oven with temperature control, bidistilled water system;
- For sample analysis: analyzer with ultra low background liquid scintillation;

Preparation of samples

In a round-bottomed flask were placed 250 mg of $Na_2S_2O_3$, 500 mg of Na_2CO_3 and 250 mL of sample. Spiked samples had a theoretical activity of 2.8070 Bq for 250 mL. Installing and starting the distillation system was made by turning agitation, heating and vacuum pump of the rotary evaporation system. It was collected 150 - 200 mL, and the last part of the distillate and the residue were discarded. 10 ml from obtained distillate were put into a counting vial, then added 10 mL scintillation cocktail.

Results and Discussion

There were prepared, measured beta spetrometric and analyzed 40 samples, divided as follows: 10 samples of distilled water and 10 spiked samples were prepared and measured by standardized method and 10 samples of distilled water and 10 spiked samples were prepared and measured by unstandardized method.

From the data obtained for the set of unspiked samples derives the following:

- results obtained by both methods are comparable and very close to the detection limit of the equipment

- in case of samples prepared by standardized method

- 60% of the samples prepared by standardized method were below the detection limit of the equipment
- measurement uncertainty was between 35.9% and 45.1%.
- average specific activity of tritium for the samples with activity above the detection limit was 0.555 Bq / L

- in case of samples prepared by of unstandardized method
  - 50 % in the samples prepared by the method of unstandardized were below the detection limit of the equipment
  - measurement uncertainty varies between 35.3% and 44.3%.
  - average specific activity of tritium for the samples with activity above the detection limit was 0.531 Bq / L

From data obtained for the set of spiked samples is noted that:
- results obtained by both methods are very close to the detection limit of the equipment
- samples prepared by standardized method
  - expanded measurement uncertainty, in percentage, varies between 3.2% and 3.3%.
  - average measured volumic activity of tritium is 11,860 Bq / L
- samples prepared by unstandardized method
  - expanded measurement uncertainty, in percentage, varies between 3.3 % and 3.6 %.
  - average measured volumic activity of tritium is 11,795 Bq / L

The distribution of the values obtained for the two methods, is graphically presented in Figure 5.3.

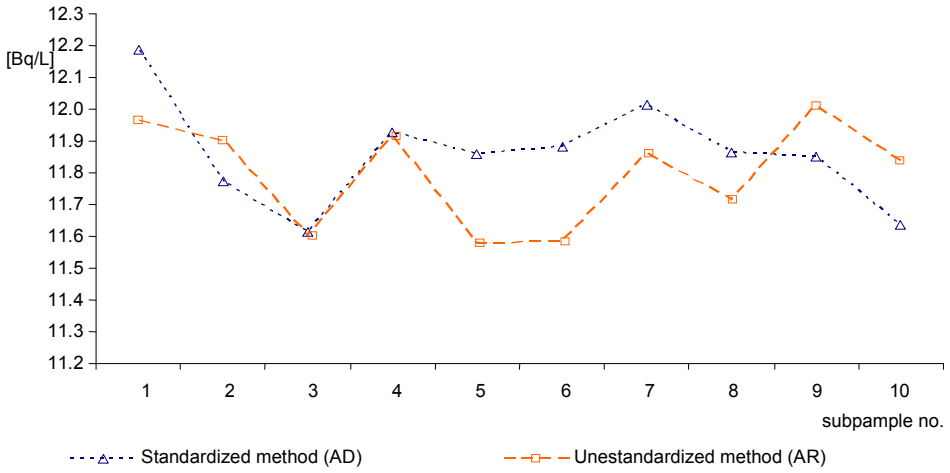

Figure 5.3 - Comparison between distribution of tritium spiked bidistilled water sample activities obtained by the two methods

In order to verify and validate the results of the unstandardized method, assessing performance criteria was done, according to ISO 9698:2010 - "Water quality - Determination of tritium activity concentration - Liquid scintillation counting method", ISO IEC 17025:2005, respectively GUM, both for the data set obtained by the standardized and unstandardized method.

Chapter 6 covers **the study of tritium dynamics in precipitations** collected from three locations in Romania. For this purpose we analyzed the results obtained in four years (2003 - 2006) of continues samples measurements, prelevated from Slobozia. Tritium concentrations were determined in precipitation compared and with values from other two locations (Craiova and Calarasi). There were calculated and analyzed the correlation coefficients between tritium concentration and the amount of average monthly rainfall collected over two years (2003 - 2004). It was analyzed the temporal evolution (monthly, seasonal and annual) of tritium activity and the influence of meteorological parameters on tritium concentrations [33], [34], [35].

Materials and Methods

Method of preparation and analysis of samples used comply with ISO 9698:2010 [90] and is presented in Chapter 5.

Samples from different laboratories were prepared and analyzed beta spectrometry at National Radioactivity Reference Laboratory from National Environmental Protection Agency.

Results and Discussion

In order to characterize the time evolution of tritium were made the following steps:

- tritium concentrations were studied over a period of 4 years (2003-2006) in precipitation samples taken from Slobozia. With this data set monthly, seasonally and yearly variations were analyzed.
- influence of meteorological parameters on tritium concentrations over 2 years (2003 and 2004) was studied in two locations: Calarasi and Craiova.
- results from Slobozia were compared with those obtained for Calarasi and Craiova, for the period 2003-2004.

In order to describe the dynamics of the atmosphere and reflect tritium trend in precipitation, samples taken between 2003-2006 from Slobozia were analyzed.

Changes in monthly multiannual

Precipitation samples were collected and analyzed individually. In order to be able to highlight the link between tritium concentration and volume of precipitation sampled, correlation coefficients multiannual related to each month were calculated.

The following criteria were used to interpret the Pearson correlation coefficient: uncorrelated (for a value that is between -0.09 and 0.09), weakly correlated (for a value that is between -0.3 and -0.1 and between 0.1 and 0.3), medium correlated (for a value that is between -0.5 and -0.3 or between 0.3 and 0.5) and strongly correlated (for a value that is between -1 0 to -0.5 or from 0.5 to 1.0) [92].

Basically, out of 12 months, in 7 months the correlation coefficient was strong, which indicates that over 58 % of the data analysis confirms the close relation between the two parameters analyzed: tritium concentration and volume of precipitation.

From the correlations values obtained for hot season it can be concluded that other factors in determining tritium concentrations could be air temperature and wind speed, especially for the months that are characterized by a reduced amount of precipitation, with high air temperatures and moderate wind speed.

For March and April were obtained weak correlation values and the values are uncorrelated in September. This can be explained by the fact that spring and autumn are generally marked by a

pronounced atmospheric instability, characterized by relatively large amounts of precipitation, significant increase in wind speed and temperature fluctuations.

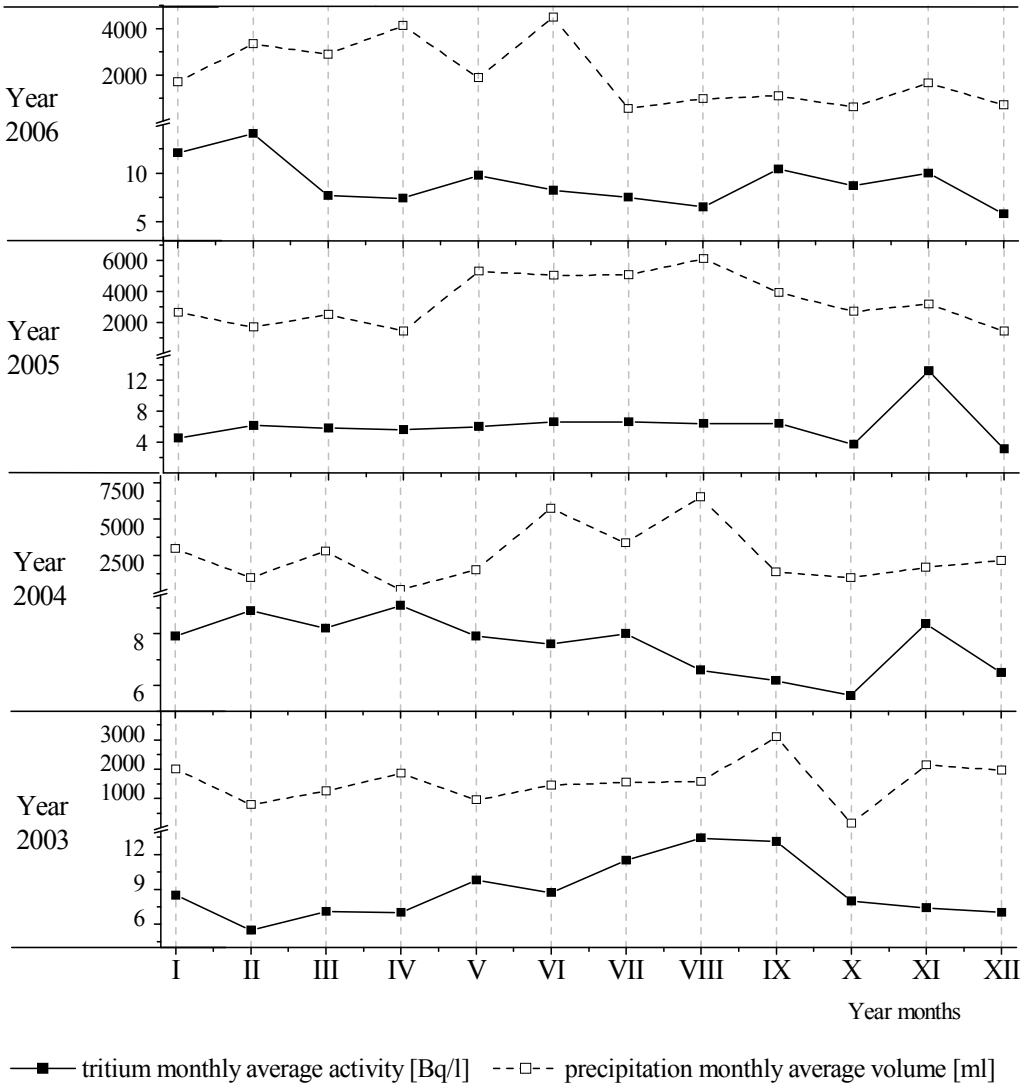

Figure 6.1 - Variation of monthly average tritium concentrations and the precipitations

It can be said that rainfall has an important role, along with other meteorological parameters of the atmosphere, the determination of concentrations of tritium.

Multi- seasonal variation of tritium

Based on data trend,one may observe an increase of tritium concentrations in small volumes of precipitation, due to prolonged periods where no precipitation occurred. In addition, large amount of rainfall leads to a decrease in the concentration of tritium (Figure 6.2).

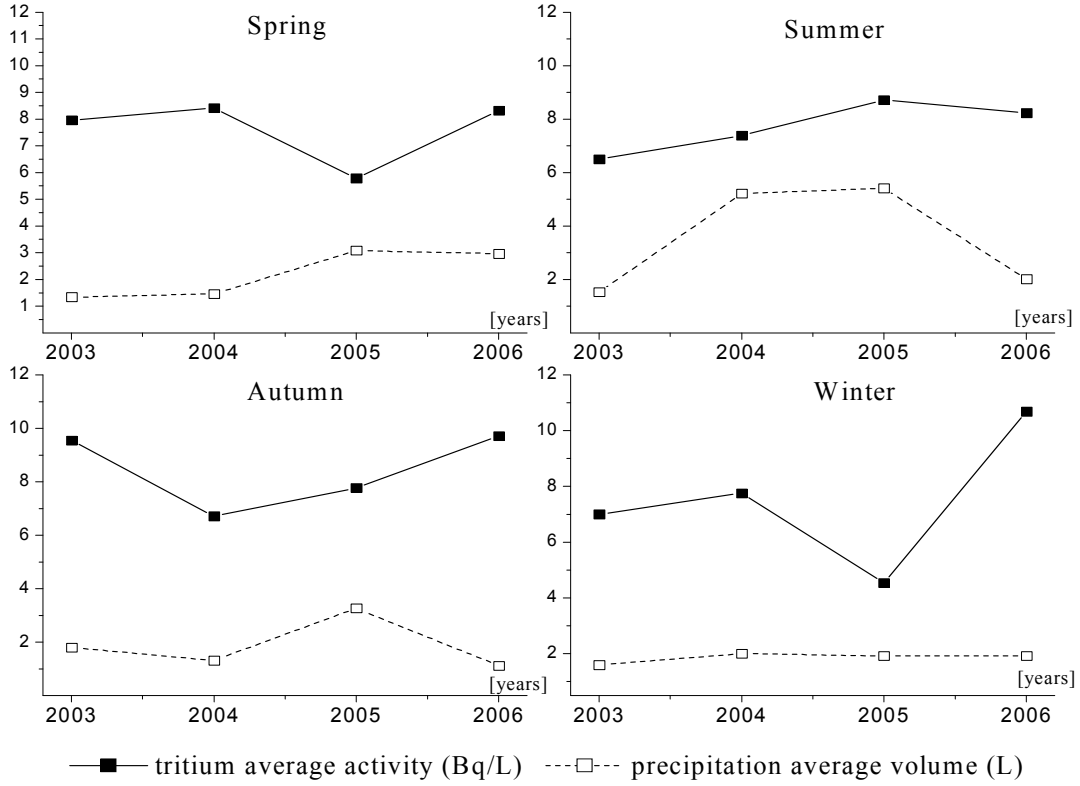

Figure 6.2 – Multi-seasonal variation of tritium concentrations in precipitations

Annual variation of tritium

Changes in atmospheric tritium, as compared with the amount of rainfall sampled over 4 years is shown in Table 6.3.

Annual distribution of observation reiterates the values shown above for the negative correlation between the amount of precipitation and concentration of tritium in the samples measured (Table 6.3). This can be explained by the values of correlation coefficients, which shows the monthly average tritium concentration dependence on rainfall, which for most months we obtained a negative value.

Table 6.3 - Annual variation of tritium concentration in precipitation

Year	**Tritium activity (Bq/L)**			**Volume of sampled precipitations (L)**		
	minimum	**average**	**maximum**	**minimum**	**average**	**maximum**
2003	5,55	8,92	13,38	0,16	1,57	3,10
2004	5,59	7,57	9,11	0,12	2,50	6,53
2005	3,05	6,15	13,16	1,42	3,42	6,13
2006	5,77	9,03	14,13	0,55	2,00	4,49

Influence of meteorological parameters on tritium concentrations in precipitation

In order to analyze the influence of meteorological parameters on the concentrations of tritium, two locations were chosen, located on the same type of relief, plain: Calarasi and Craiova. The differences between the two sampling points chosen consist in the distance from the Black Sea whose currents affect the town Calarasi, but with no effect in Craiova.

The meteorological parameters used in this chapter were taken from the web - site www.freemeteo.com [93].

From the data presented in figure 6.3 it is noted that although the data are consistent, specific locations particularities appear, leading ultimately to a trend in tritium concentrations that is characteristic for each location.

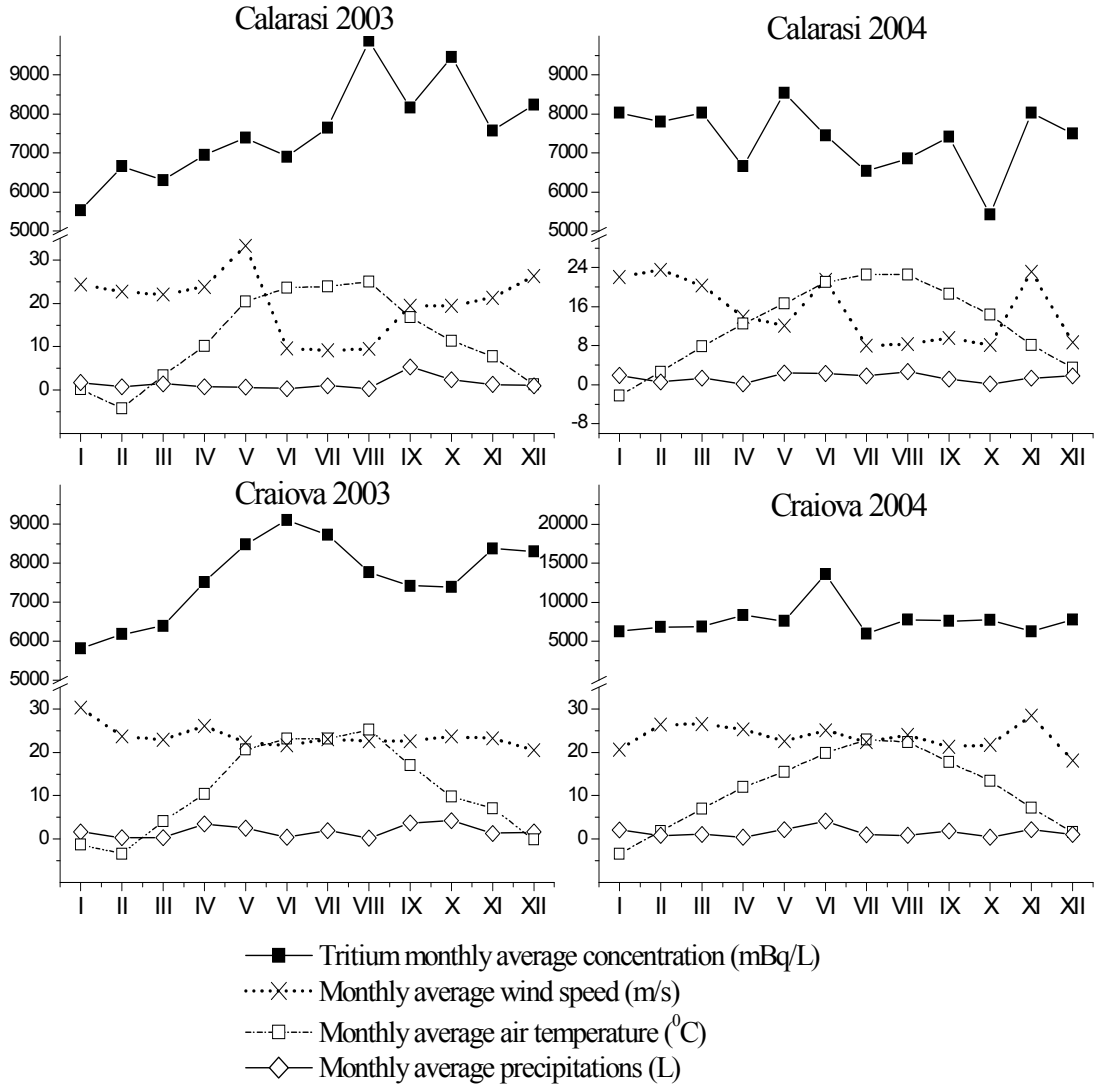

Figure 6.3 - Average annual variations in tritium concentrations in precipitation

It was calculated and presented analysis of the correlation between monthly average weather parameters and monthly average concentration of tritium in precipitation, depending on the sampling site. The values obtained indicates that the atmospheric concentration of tritium at a time is equally influenced by meteorological parameters analyzed.

Analysis of the data presented indicates that Slobozia and Calarasi locations (at approx. 40 km in a straight line one from another and approx. 59 km, respectively 58km in a straight line to Cernavodă [94]) were not affected by nuclear power plant (NPP) activity, having values in the same range as those registred in Craiova (located at approx. 336 km, in a straight line, from Cernavodă [94]), respectively in the natural background for tritium in precipitation. Following the above, it can be concluded that Slobozia and Calarasi can be used as control points for monitoring the operation of Cernavoda NPP [95].

Table 6.6 - Correlation between monthly average weather parameters and annual average concentration of tritium in precipitation

Year	Sampling location	Correlation with temperature	Correlation with wind speed	Correlation with precipitations
2003	Călăraşi	0,475	-0,334	0,126
	Craiova	**0,685**	**-0,645**	0,062
2004	Călăraşi	-0,410	**0,589**	0,425
	Craiova	0,365	0,071	**0,650**

Choosing the three sampling points in the same relief structure, the Romanian Plain, allowed analysis of tritium dynamics from precipitation and also demonstrated the fact that meteorological parameters have a significant influence over the tritium concentration in precipitation.

Determination of tritium specific activity in soil is presented in **Chapter 7**. The case study in this chapter presents an interdisciplinary analysis of soil samples taken from the arboretum of the Institute of Physics and Nuclear Engineering " Horia Hulubei " (IFIN -HH), located about 2 km from the institute and a few kilometers from the city Bucharest.

Three areas of the park were selected in order to be investigated. The choice of sampling points was based on the following considerations, which combines sampling criteria of three areas: history, geology / geophysic and nuclear:

- F1 sampling point was chosen in the open field, at the base of the present stairs to Castle Otetelesanu, which corresponds to an open area without trees and covered with spontaneous vegetation, growing at the foothills of the hill leading up to the castle. According to data held by us, the land has maintained this about 160 years.
- F2 sampling point was chosen in a semi-enclosed where secular trees grows on the shores of the interior lake, which is characterized by low vegetation on the ground and sporadic presence of trees.
- F3 sampling point was chosen in an area of lush and dense vegetation on the ground, located in forest area defined by growth of symbiotic species of native and exotic trees, located in proximity to one of the lake's water channels.

The three areas are characterized by a layer of earth fill, of variable thickness according to the depth from the bottom of the swamp, which has been added in the moment when the water was drained.

Materials and Methods

Samples were brought in the laboratory and prepared in order to determine the tritium specific activity. So, the first step was to determine the moisture content for each subsample, using 2 g. In parallel, the sub-samples were subject to extraction of water content from soil pores, by using the azeotropic mixture of toluene - water [31], [98], [99], [100].

Water samples of the same subsamples were mixed with scintillation cocktail in 20 mL polypropylene measurement vials, with teflon caps (model Packard) and measured by beta spectrometry.

Measurements were made using beta spectrometry by liquid scintillation and the technic of results intercomparison on three different spectrometers. In addition, for validation of results two kinds of scintillation mixtures were used (Instant Gel Plus, in combination 8:12 (v: v) and Ultima Gold LLT in combination 10:10 (v:v).

Spectrometric analyses were performed in two laboratories as follows: Radiochemistry Laboratory for Environmental Samples (LRPM) of IFIN -HH, using a Tri- Carb, model 1600 TR and the National Reference Laboratory for Radioactivity (LNRR) of the National Environmental Protection Agency (NEPA) using a Tri- Carb, model 2770 SL / TR and Quantulus, model 1220.

Results and Discussion

Sampling was limited to the groundwater level because, due to drilling equipment used, the contamination probability of the lower layer of water from the above area was high, which would made the analysis to become irelevant.

Over 77% of the values obtained with Tri -Carb 1600 TR, when using Insta-Gel Plus as scintillation liquid, were placed below the detection limit of the equipment.

For measurements on Tri -Carb 2770 TR / SL, using Ultima Gold scintillation cocktail, values were placed over the LD, but the combined standard uncertainty for k = 1 [90], [101], [102] exceeded the average value of 20 %.

The measured results using the equipment Quantulus 1220 were much better than those made with other spectrometers, since they take into account all the errors introduced in the measurement phase.

From data analysis it is noted that most of the values readings made using Ultima Gold LLT were above the equipment's detection limit, with good counting statistics and is not required electrolytic enrichment nor counting at very low levels of radioactivity, which certify once more the reliability of this type of cocktail for measuring samples with low activities.

Results led to the conclusion that the use of Insta Gel Plus is not suitable for determining low activities in samples.

Variation of tritium specific activity over depth in the soil to the groundwater level in the three sampling points in the arboretum of IFIN -HH is plotted in Figure 7.2.

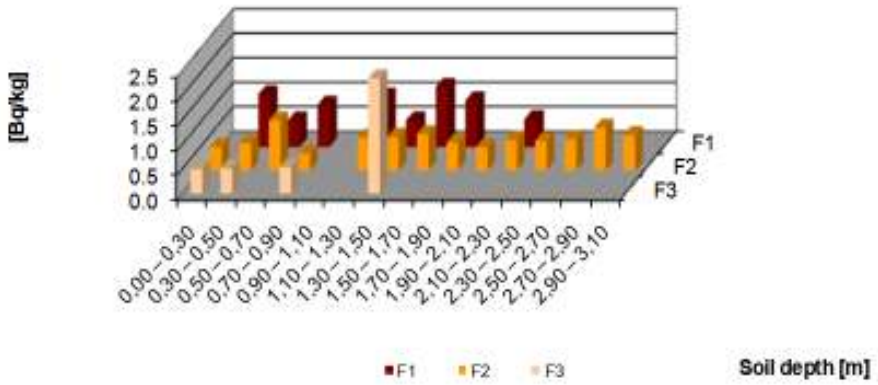

Figure 7.2 - Variation of tritium specific activity over depth in the soil in the three boreholes from IFIN- HH arboretum.

The graphical representation of the variation in the specific activity of the tritium in the ground at various depths is shown in Figures 7.3, 7.4 and 7.5 depending on the absolute humidity.

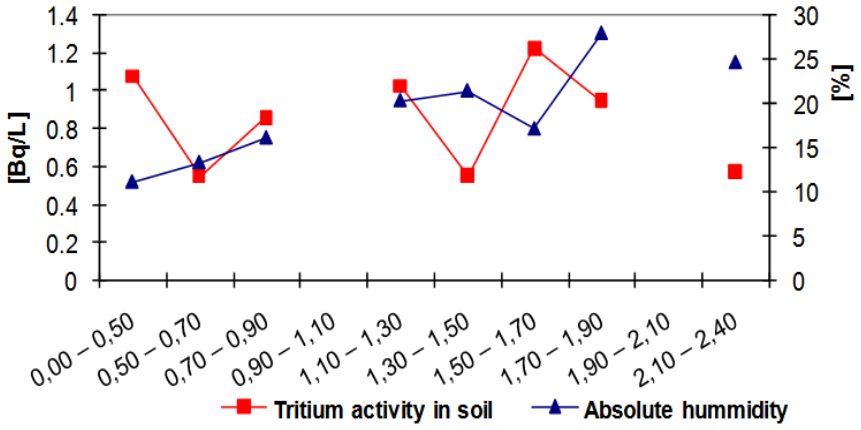

Figure 7.3 - Variation of tritium specific activity in soil drilling F1 at different depths depending on the moisture

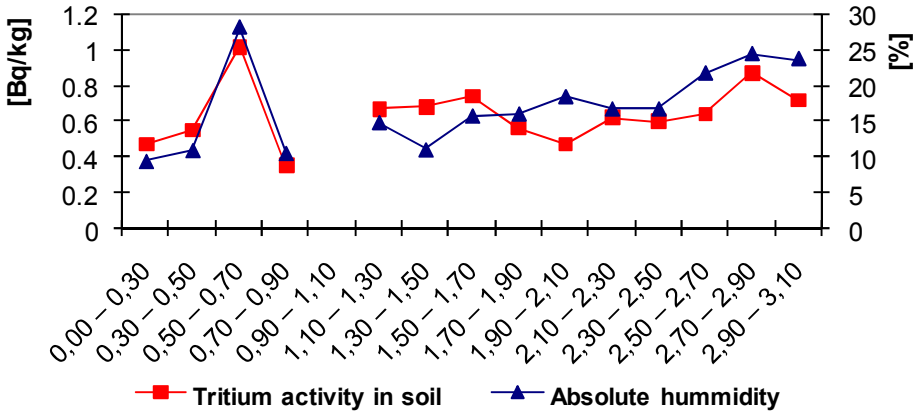

Figure 7.4 - Variation of tritium specific activity in soil drilling F2 at various depths, depending on the moisture

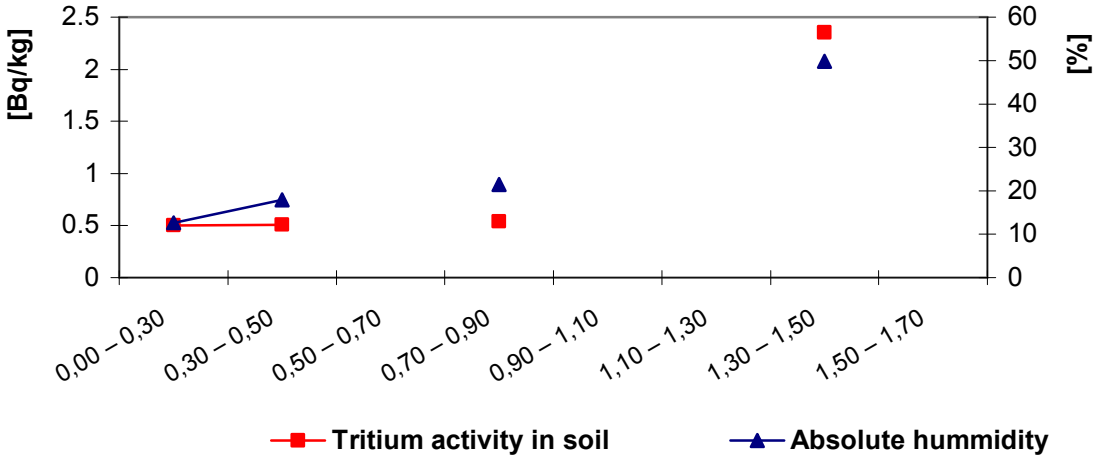

Figure 7.5 - Variation of the specific activity of the tritium in the drilling F3 soil at different depths, depending on the moisture

Presence of the maximum recorded value in drilling F3, located in the forest, compared with F1, which is located close to the building which was used as a laboratory, clearly indicates that tritium found is mostly due to the current activity conducted by IFIN -HH.

Analysing tritium specific activity obtained for the three boreholes are the following aspects:

• same general tritium accumulation trends in all analyzed boreholes profiles: the first portion associated with stuffing soil, there is a surface accumulation, after which it drops dramatically near the clay layer; followed by maximum accumulation is the first portion of clay layer (except F3), followed by a first zone in which the amount of tritium is decreasing visible; it continues with periodic and variable concentration clay layer and it ends up by a constant decreasing in a layer of sandy clay, which represents the swamp's end and beginnig of the groundwater layer;

• there is a difference in height between the three wells in succession above mentioned areas;

• there is a valoric difference between boreholes, F1 is the most affected by the presence of tritium in the atmosphere and precipitation, folowed by F2 and the last, F3, is the most protected;

• the accumulation of tritium between 150-170 cm depth in all three boreholes, at the top of the clay layer; for F3, this section is adjacent to the layer of sandy clay and is the last of the clay profile;

• from the F1 and F2 boreholes data one can see that the accumulation is fixed on the top muddy clay layer, so the slight contamination is coming from infiltrated surface water and not from groundwater, as it appears form F3;

• profiles are following very well the distribution of water in soil, due to both lithologic profile, as well as the distribution pattern of the plants and trees roots in depth;

• borehole profiles, in Bq/L water are not identical to those presented in Bq/kg soil. The first ones expresses better the properties of water retention in the soil pores, as compared to the second set, in which only the activity contained in one kg of soil at a given moment, depending on the pluviometric regime at sampling time.

The case study presented in **Chapter 8** contains the **monitoring of the environment in the event of a nuclear accident or radiological emergency, Fukushima case.** This chapter analyzes the variation of artificial radionuclide concentration, ^{131}I, in atmospheric aerosol samples and atmospheric deposition, taken by Environmental Radioactivity Surveillance Stations, alongside with the monitoring strategy applied.

On 11 March 2011, in Japan, Dai- ichi NPP from Fukushima Prefecture had a major nuclear accident, rated at level 7 on the INES scale (after the Nuclear and Industrial Safety Agency (NISA)).

Rated as the second severe accident, after Chernobyl, having civil consequences, it released in the atmosphere between (1.3-1.6) • 10^{17} Bq ^{131}I and (6.1 to 15) • 10^{15} Bq ^{137}Cs [106]. Particles containing 131I and 137Cs arrived in a few weeks the entire northern hemisphere. Many laboratories around the world have mobilized and detected and informed the authorities and public opinion on the measured concentrations within their national territories. Also, meteorological institutes (Meteo France) published maps with the estimated dispersion of the radioactive cloud.

Environmental radioactivity monitoring in Romania is conducted by the National Environmental Radioactivity Surveillance Network (NERSN) under the Ministry of Environment and Climate Change. NERSN continuously monitored the atmospheric dispersion of pollutants through atmospheric aerosols and atmospheric deposition.

The case study pesented in this chapter is analizing the monitoring strategy used for detection and tracking of pollutant plume, originated from the accident at the Dai-ichi NPP, Fukushima, in Romania [108], [109], particularised on artificial radionuclide ^{131}I.

Materials and Methods

Air quality in terms of radioactivity in Romania is continuously monitored by NERSN, under the Standard Monitoring Programe (PSM), throughout gamma dose rate measurements (hourly), total beta analyses of atmospheric aerosols and total atmospheric deposition (wet and dry), for screening purpose as well as qualitative and quantitative gamma spectrometric analysis for detection of radionuclides present in the sample.

Location methodology, sampling and analysis is unified in NERSN and aims to ensure consistency of information provided in a given time.

Sampling points of atmospheric aerosols and atmospheric deposition are permanent, while ensuring data traceability.

Choosing the location for sampling devices was based on a unique methodology, which provides minimum requirements and mandatory:

- Location of sampling devices outside urban areas, away from pollution sources (road traffic, baskets, furnaces, etc.).
- Minimum distance between the device and other objects / obstacles (buildings, fences, trees, etc.) from terrain must be twice the height of the obstacle.
- Presence of luxuriant vegetation is not allowed.

Sampling of atmospheric aerosols was carried out on glass fiber filters, with a retention factor of 98 %, located at 2 m above the ground and connected to a vacuum pump, with a sampling flow rate of 5 m3 / h. The sampling period was of 5 hours between the hours 02:07 (A1), 08:13 (A2), 14:19 (A3), 20:01 (A4). Laboratories with 24-hour work schedule performed all four samplings and laboratories with 11 hour work schedule performed only the first two samplings.

Total atmospheric deposition sampling was performed with atmospheric deposition collectors, having an interior polypropylene covered and an area of 0.3 m^2. The sampling period was of 24 hours and the daily sampling time was 9 am.

For gamma spectrometric analyses it was used a NERSN's network of high purity and resolution broad germanium gamma spectrometers (HPGe), from Canberra, model BE 3820, covering a range from 3 keV to 3MeV with nominal efficiency of 35%. Energetic resolution at 5.9 keV was 450 eV, at 22 keV of 750 eV and at 1332.5 keV of 2100 keV. The Genie 2000 software was used for analysis. Energy calibration was performed using spectrum's peaks and efficincy calibration was performed by Canberra's ISOCS. Efficiency calibration verification was performed using reference materials from IAEA. Corrections for geometry, density, matrix and summation were performed.

Results and Discussion

For investigation's purpose there were taken into account the following assumptions: great distance between the place of the accident and Romania (8000 km), evolution of the weather condition at that time, direction and height of air currents and also the pollutant plume predictions worldwide by various national and international organizations ability and not least the results reported by countries in the way of cloud, availability of existing technical and methodological framework NERSN.

Hourly monitoring of gamma dose rate using automatic stations, total beta measurement of immediate atmospheric aerosols and total atmospheric deposition revealed no detectable variations of natural background radiation.

Environmental pollutant dispersion was large enough not to be evidenced by global beta tests, requiring a thorough investigation, on time insterval as short as possible. Therefore gamma spectrometric analysis was used for short periods of time, which ranged from 24 hours to several days.

In the NERSN are 37 Environmental Radioactivity Surveillance Stations (SSRM) that provides continuous sampling of the samples, which are then analyzed by gamma spectrometry network comprising six high-resolution gamma spectrometers located at Bucharest, Craiova, Constanta, Iasi, Arad, Baia Mare. In normal situations, acording to PSM, gamma spectrometric analyses are performed on monthly cumultated, with daily sampling.

Given the relatively short half-life of ^{131}I (8.0233 ± 19 days [15] and small activities targeted, the measurement strategy has been amended accordingly [108]:

- Minimum alocated measuring time for a single measurement was of 40000s, being large enough to detect activities $x10^{-3}$Bq,
- For atmospheric aerosols priority samples were collected by the Environmental Radioactivity Surveillance Stations (SSRM) in the mountains, because, given the distance from the point of emission, the pollutant cloud was already entrained by high altitude air currents; followed by gamma spectrometry laboratories that performed the analysis and then the rest of SSRM.
- For atmospheric deposition, priority samples were the ones taken from the locations where have been registered wet deposition (precipitation).

Location point of sampling at 1850 m allowed to analyze the contamination level of high altitude air currents, who are responsible for transportation and global scale atmospheric dispersion of radionuclides released into the atmosphere as a result of the Fukushima accident.

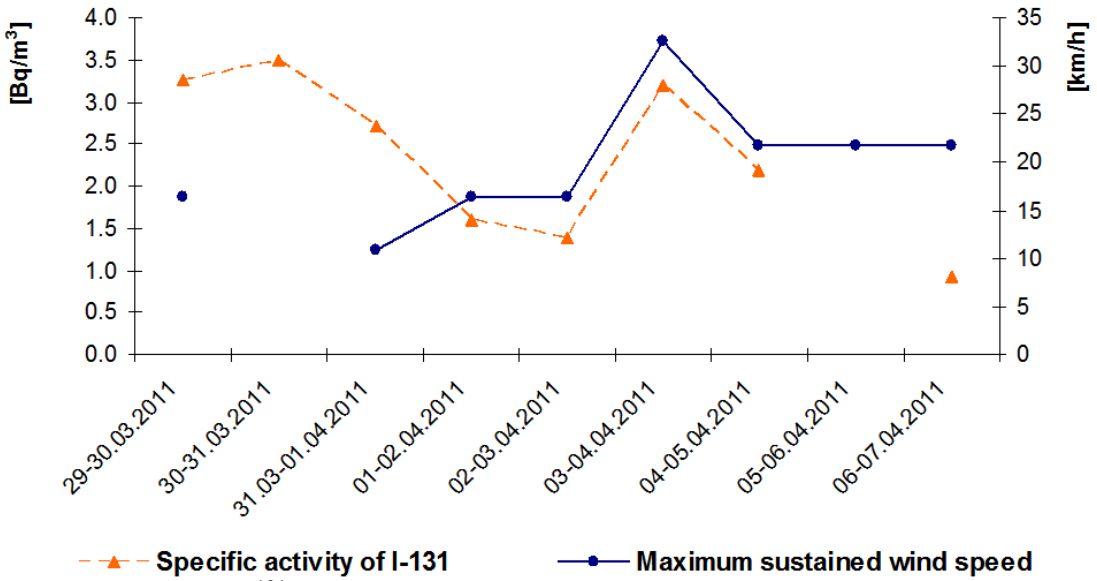

Figure 8.1 - Variation of ^{131}I activity in atmospheric aerosols samples collected by SSRM Toaca, between 29.03-07.04.2011, compared with the maximum wind speed [93]

^{131}I maximum concentration in atmospheric aerosols samples was registred in 30 - 31.04.2011, for a combined sample consisting of four filters, taken from Toaca Peak of Ceahlău Mountain and was 3,494 ± 0,247 mBq/m^3.

As expected, data presented in Figure 8.1 shows the presence of a correlation between the specific activity of 131I in atmospheric aerosols and wind speed during sampling. Value resulting from the Pearson correlation is -0.240, idicând that they are poorly correlated. The reason being the concomitent appearance of other weather phenomena, in paralel or intermitent with air currents speed, such as fog, indicating stationary atmospheric conditions.

From the information presented regarding that intensively monitored period it was noted that the site was under the radioactive contaminated cloud for two weeks, between 29.03.2011 - 07.04.2011. During this period, the recorded values were in the range between 0.925 ± 0.107mBq/m^3 (recorded during 06 - 07.04.2011) and 3.257 ± 0.260 mBq/m^3 (recorded during 29 - 30.03.2011).

It is noted that the beginning and end of the period in which the presence of ^{131}I in atmospheric aerosols was revealed is clearly bounded by the extreme values recorded. Except intermediate maximum recorded within the range 03 - 04.04.2011 (3.210 ± 0.200 mBq/m^3) is a decreasing trend prevailing.

In Table 8.2 are shown ^{131}I specific activity values along with the locations in which the atmospheric aerosols samples contaminated with ^{131}I were taken during March-April 2011.

Due to very low concentrations existing in the atmosphere, only in four locations were revealed concentrations above the detection limit of the equipment, in cumulated samples for a short sampling interval (24 - 72 h). In the remaining cases was highlighted artificial radionuclide ^{131}I in monthly cumulated samples.

As can be seen from Figure 8.3 the specific activity values recorded for ^{131}I are directly correlated with altitude up to 1850 m, above this altitude, the obtained values were below the limit of detection (SSRM Babele, 2500 m). A possible cause of this phenomenon may be the position of the two sampling points in relation with the air curents direction.

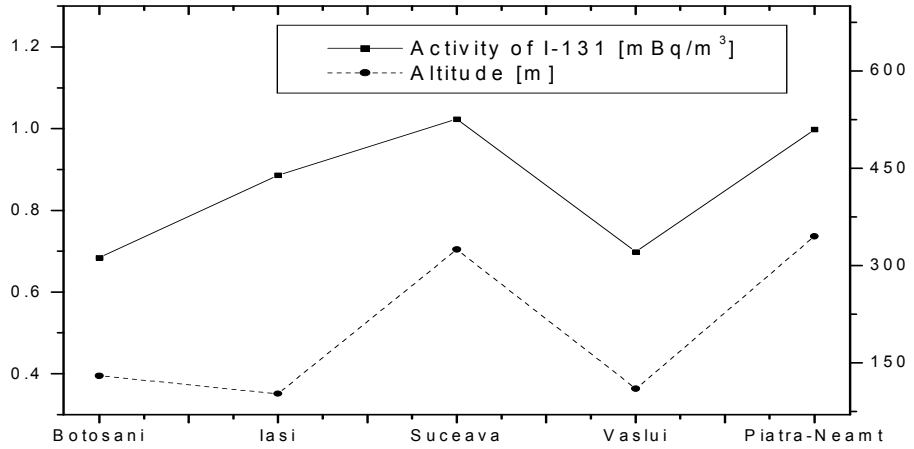

Figure 8.3 - Changes in ^{131}I concentration from atmospheric aerosols, according to altitude

By analyzing only the values above the equipment's detections limit, one can see that they falls between 0.225 ± 0.036 mBq/m^3 (recorded during 26-27.03.2011 at Constanta) and 2.300 ± 0.110 mBq/m^3 (registered in during 03-04.04.2011 in Constanta).

Registration of both high and low values in the same location is explained by the fact that, at national level, the direction of air curents, during the monitored period, was from WNW to ESE. Also, both the presence and the Carpathians form plays an important role in deviating the air curents from their original route, thus justifying why most locations where ^{131}I has been identified were in the NE of Moldova and Dobrogea (Constanta) respectively.

Taking in the consideration the sampling period, besides the location, it is noted that the spatio-temporal distribution of atmospheric aerosol samples reveals the presence of ^{131}I, artificial radionuclide, originated from the nuclear accident at Fukushima, in March, beeing mainly concentrated in the NE part of the country, while in April there was a slight shift to the center.

In addition, the number of cumulated samples in a short time period, in which ^{131}I have been identified, halved in April, and monthly cumulative number of samples increased by 20 % from the previous month, indicating a decrease in the activity of iodine dispersing into the atmosphere.

For samples of atmospheric deposition, ^{131}I is observed only when wet deposition (precipitation) have occured, due to washing phenomena of the radioactive cloud over the sampling localities.

Radioiodine absence in some precipitation is explained by the fact that the two moments do not coincide in time, the radioactive cloud was not over localities when it rained.

^{131}I values were first recorded on 27.03.2013, in many location in the country, mainly located in the eastern half. ^{131}I was detected in deposition samples before the atmospheric aerosol samples. This leads to the conclusion that atmospheric deposition samples are better indicators than atmospheric aerosol samples.

Maximum ^{131}I concentration was 3.572 ± 0.127 Bq/m^2 day (Figure 8.4) and was recorded in Constanta, in the sampling interval between 02-03.04.2011, the explanation being given by the weather conditions in this area, which generate abundant rainfall. The value obtained in Constanta correlated with the absence of any values above the detection limit of the equipment in other parts of the country, during the same period, suggests that the cloud came to Constanta from the seaward (Black Sea) and was blocked for a period of time over this area. This may be associated with a

second wave of the radioactive cloud penetration in our country, which after a while stationed above the Black Sea has dispersed in the coming days almost everywhere in the country (07-09.04.2013) when values above detection limits were found in atmospheric deposition of several sampling points. We conclude that the country was traversed at least twice by the radioactive cloud, the first time around 27.03.2011 and again between 02-03.04.2011.

From the data presented it is noted that the majority of individual atmospheric deposition samples for which was identified the presence of ^{131}I were taken from the south and center of the country.

Comparing the time periods in which ^{131}I have been identified in samples of atmospheric aerosols, with the ones from in atmospheric deposition samples one can see that they complement each other, creating an overview of the period in which the radioactive contamination cloud passed over each location.

The last three samples of atmospheric deposition in which iodine has been found were taken after intermittent periods of rainfall, extended over several days, which confirmed that the sampling point was not always under the radioactive cloud.

In figures 8.7 and 8.8 is presented the dependency of the specific activity of the 131I artificial radionuclide, coming from Fukushima, in atmospheric deposition samples, with the amount of rainfall sampled in 24 hours, the sampling time, as well as the altitude of sampling point.

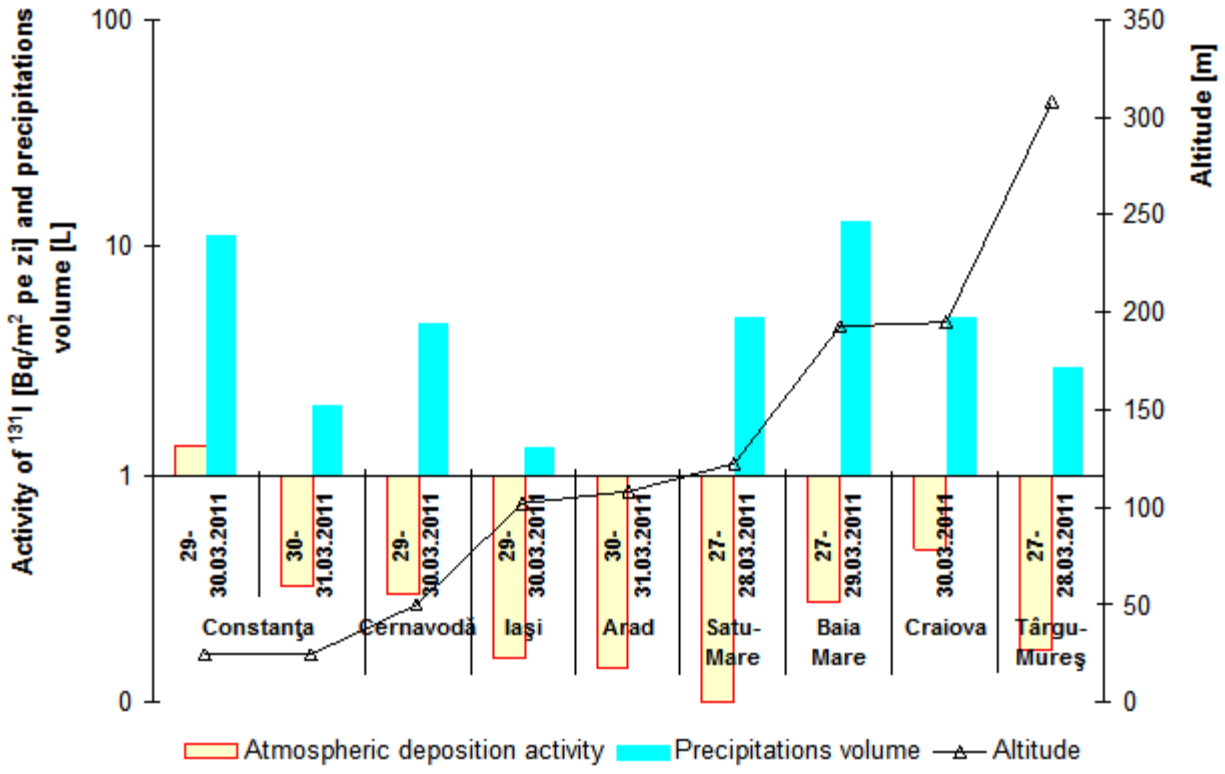

Figure 8.7 - Detection of ^{131}I in atmospheric deposition - March 2011

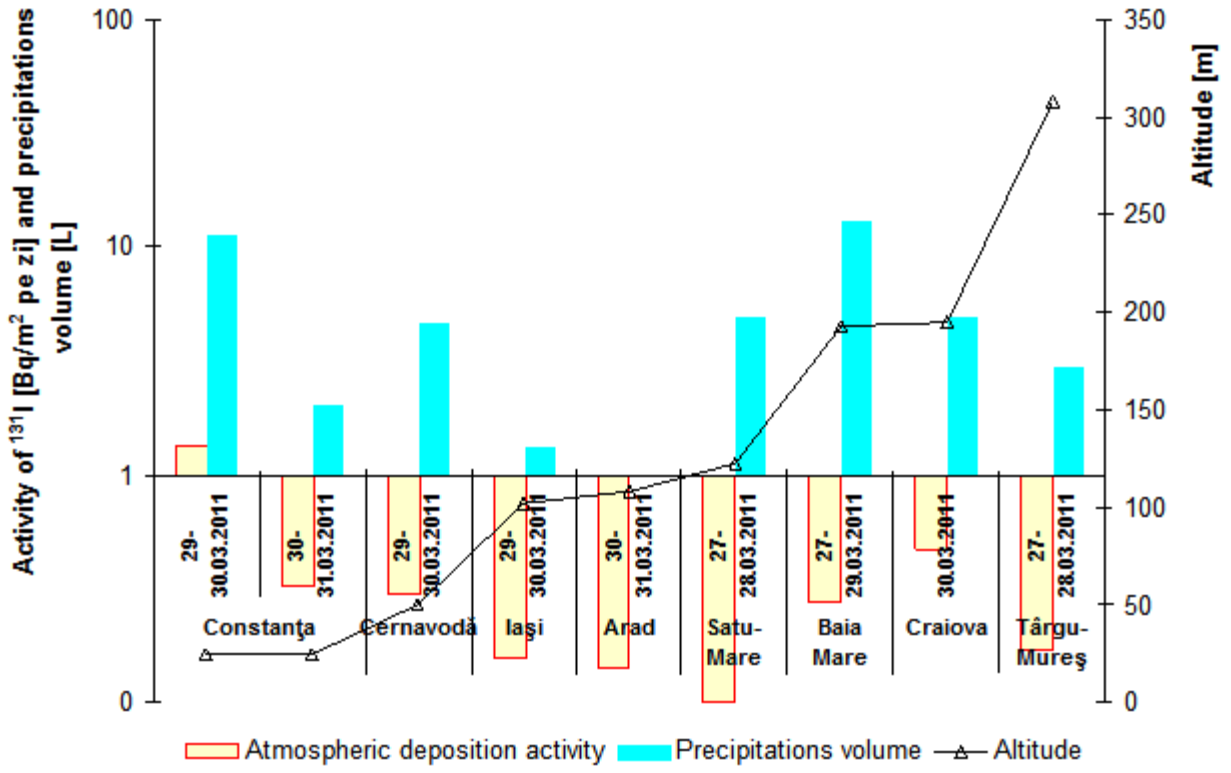

Figure 8.8 - Detection of ^{131}I in atmospheric deposition - April 2011

Chapter 9 is dedicated to **the study of correlation between atmospherical radioactivity and meteorological variables**. Monthly average variation of atmospherical deposition samples, atmospheric aerosols samples and gamma dose rate, collected over one year by the Environmental Radioactivity Surveillance Station Botosan, were analyzed depending on weather variables (atmospheric precipitations, atmospheric pressure, wind speed, air temperature and relative humidity).

Natural radiation sources provide for 85.5% of the average radiation dose received by the population and are mainly due to cosmic radiation, terrestrial radiation and radon [3]. Radon is an noble gas, which is emitted permanently by soil, in various rates, depending on the soil type. The three radioactive isotopes of radon ^{222}Rn (half-life of 3.824 days), ^{220}Rn (half-life of 55.6 s) and ^{219}Rn (half-life of 3.96 s), occur in the decay chains of 238U, 232Th and 235U, respectively, existing in traces in the Earth's crust [9]. The population exposure to the radioactive element radon is now regarded as the most significant single contribution to human irradiation by natural sources [3], [18].

The weather conditions influence the level of radiation which is directly related to the people health. This paper presents the effects of meteorological variables on atmospheric natural radioactivity, using gamma dose rate and total beta activity of atmospheric aerosols and total depositions.

Materials and Methods

Values shown are monthly averages achieved throughout the year from measurements performed on environmental samples by Environmental Radioactivity Surveillance Station (SSRM) from Botosani, Romania.

The principle of measuring the total beta activity of the atmospheric aerosol is based on the samplingof the aerosol on glass fiber filters with a high coefficient of retention (96 - 99 %). The suction head is attached to 2 m above the ground and is connected to an aerosol sampler.

Sampling time was 5 hours, four times a day as follows: 02:00 to 7:00 (aspiration A1), 08:00 to 13:00 (aspiration A2), 14:00 to 19:00 (aspiration A3) and 20:00 to 01:00 (aspiration A4). The filters were measured after 3 minutes from sampling, for 1000s, after 20 hours, for 3000s and after 5 days, for 3000s [118], [119]. The first two measurements provide information necessary to determine the concentrations of radon and thoron progeny and the last measurement is used to identify the presence of artificial radioactivity into the atmosphere. Errors for dose rate, the efficiency of the filter and the etalon source have a maximum estimated value of 20 %

Total atmospheric deposition samples were obtained by collecting the dust and daily rainfall using a standard collector located at 1 m above the ground. The standard collector was coated inside with a layer of polyethylene.

Sampling time was 24 hours, with a daily frequency, measured daily to determine the immediate total beta activity, for 1000 seconds.

Table 9.2 - Distribution of monthly weather variables

Month	Meteorological variables				
	Precipitations (l)	Relative humidity (%)	Pressure (hPa)	Temperaturea (^{0}C)	Wind speed (m/s)
January	0.33	92.61	1008.8	-6.89	2.70
February	0.21	93.45	992.6	-6.23	2.52
March	0.30	93.42	1004.1	-2.74	2.29
April	0.65	75.87	1001.0	9.43	3.24
May	0.29	71.20	996.5	18.77	4.00
June	0.63	67.33	1001.9	20.26	2.68
July	0.81	79.00	1000.5	19.12	2.52
August	0.64	81.06	1001.3	18.72	2.19
September	1.24	87.17	995.4	11.82	3.15
October	0.34	87.90	1003.3	9.46	2.39
November	0.57	88.63	999.8	7.27	2.06
December	0.72	96.29	1002.5	-3.99	1.79

In order to determine the dependence of atmospheric natural radioactivity on meteorological variables, Pearson's correlation coefficient [120] was used. Pearson correlation coefficient quantifies the degree of dependence of one physical measure of another physical measure, giving a value between +1 and −1 inclusive. The sign of the correlation coefficient indicates the relationship between the variation of two physical measures: positive values indicate that when the values for one set increases, the values for the other set also increase; negative values indicate that when the values for one set increase, values for other set decrease; values near zero means that there is a random, nonlinear relationship between the two set of variables.

For interpretation reasons, the following criteria was used for Pearson's correlation coefficient: uncorrelated (for a value that is between -0.09 and 0.09), low correlated (for a value that is between -0.3 and -0.1 or between 01 and 0.3), medium correlated (for a value that is between -0.5 and -0.3 or between 0.3 and 0.5) and strong correlated (for a value that is between -1.0 and -0.5 or between 0.5 and 1.0) [92]. The Person correlation coefficients were computed by means of Microsoft Office Excel using the function “PEARSON”.

Results and Discussion

The variation in monthly average values of meteorological variables (atmospheric precipitations, atmospheric pressure, wind speed, air temperature and relative humidity) and atmospheric deposition, atmospheric aerosols and total gamma dose rate are presented in Figure 9.1.

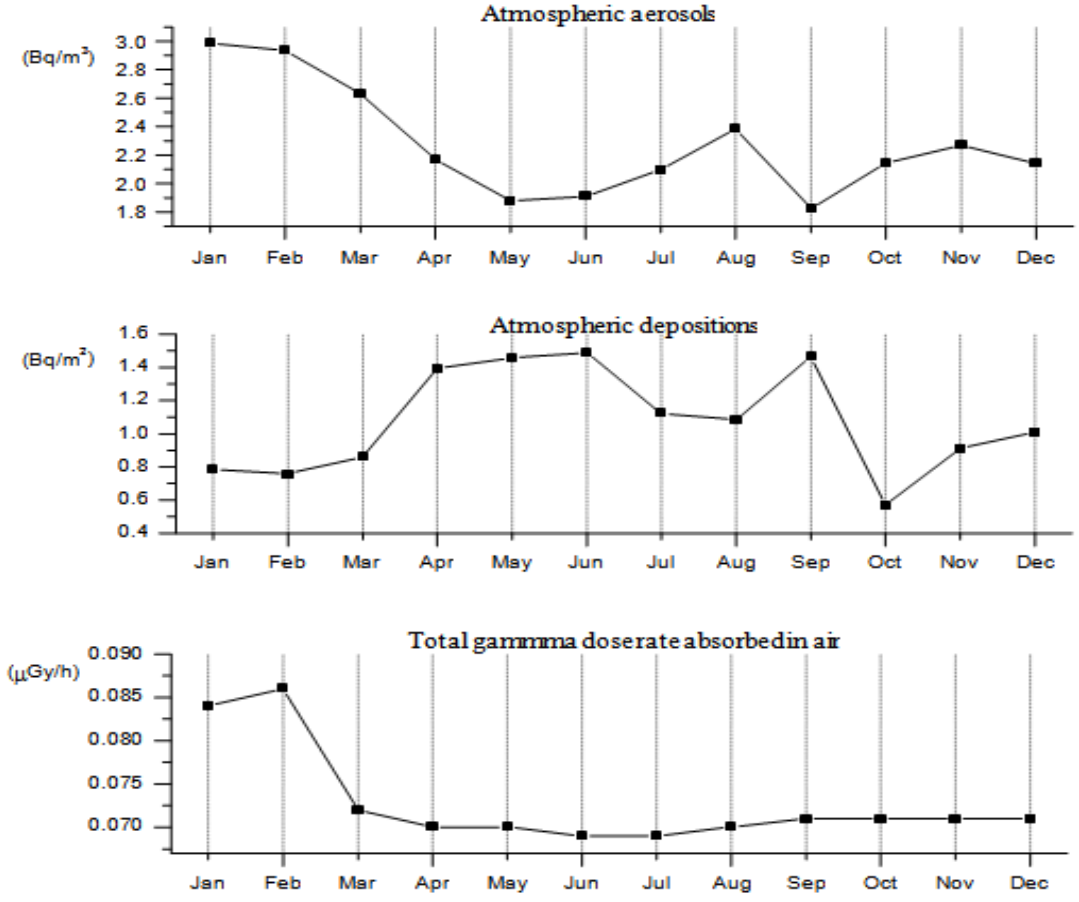

Figure 9.1 - Monthly distribution of radioactivity in air

In case of winter months it can be seen that the precipitations, air temperature and wind speed have low values, while the total gamma dose rate and total beta activity concentrations for atmospheric aerosol have maximum values due to atmospheric stability, consequently the total beta activity concentrations for atmospheric deposition are minimum, due to the fact that the precipitations are minimum.

In summer months, when the high temperatures occurs, the minimum values are obtained for the total gamma dose rate and for atmospheric aerosols concentrations due to dominant convection air currents, combinet with low wind speed.

The minimum value for total beta measurements on atmospheric aerosols and the maximum value for total beta measurements on atmospheric deposition from autumn, especially from September, can be explained by the maximum values obtained for precipitations (having as result a dust free atmosphere), an increase of wind speed (dominant factor for dust transport) and low values for relative humidity and atmospheric pressure.

From correlation analysis of monthly averages of atmospheric aerosols total beta activity concentration with the meteorological variables it was found that they are well correlated with air temperature and wind speed, both factor in the movement of air masses [92], [122]. From the same table it can be seen that the relative humidity, atmospheric pressure and rainfall are generally weak global beta activity correlated with atmospheric aerosols.

Regarding the correlation with atmospheric precipitation, October is the only month in which the medium correlation was registered, this is justified by the greater amount of precipitation recorded, compared with the other months of the year, and by the fact that it was the month in which occurred the most days of consecutive atmospheric precipitations.

The link between monthly averages of global beta activity concentration of total atmospheric deposition and meteorological variables is presented by analyzing the correlations between the two parameters, showing a good correlation with rainfall and relative humidity, both directly influencing deposition rates of dust particles in the atmosphere on the ground. Atmospheric deposition are poorly correlated with air temperature and atmospheric pressure and generally uncorrelated with wind speed.

From the correlation analysis it can be clearly seen the influence of weather conditions upon atmospheric natural radioactivity. The most influent variables on the natural radioactivity are air temperature and relative humidity; wind speed and precipitations have moderate influence and atmospheric pressure is less influent.

From the analysis of correlation coefficients between monthly averages of gamma dose rate absorbed in air and meteorological variables, it was found a strong correlation with air temperature and relative humidity, rainfall average correlation because of the quantity of rainfall (washing of atmosphere) an micro-atmospheric deposition layer is deposited on the surface of the detector (leading to a small increase of the results obtained), while the pressure and wind speed are poorly correlated.

Chapter 10 presents **applications of multiple linear regression model for estimating and predicting progeny concentrations of ^{222}Rn and ^{220}Rn**, using concentrations and meteorological variables values from Botosani laboratory. In order to implement the model were studied collinearity by calculating the Pearson correlation coefficient and level of significance, and multicolinearity by calculating the variance inflation factor.

To achieve analytical expressions regression equations were used only current variables. Performance model for predicting progeny concentrations were revealed using the correlation coefficient between the measured values and those generated by multiple linear regression model.

Chemically inert gas, radon (^{222}Rn) and thoron (^{220}Rn) are decay products of ^{226}Ra and ^{224}Ra, elements still of natural decay series, headed by ^{238}U series and ^{234}Th respectively, which are present in all terrestrial materials. Due to high toxicity, determining their concentration is an important concern for radioprotection and environmental specialists.

In view of the exhalation from the ground, radioactive gases ^{222}Rn, ^{220}Rn and its progeny (daughters) enters in the atmosphere, where their concentration is influenced by the dynamics of the specific weather conditions of planetary boundary layer: air temperature (Ta) atmospheric pressure (Pa), wind speed (V), precipitation (Pr) and relative humidity (U_{rel}) [123], [124], [125], [126], [127].

The main purpose of this chapter was to complete the missing values for the indicators of interest from data series, in order to characterize the environmental radioactivity. For the multiple linear regression model was applied on a set of historical data, validated.

Colinearity and multicolinearity were analyzed for independent variables (predictors) in order to obtain reliable results by applying multiple linear regression model.

The colinearity study was based on analysis of the correlation matrix of predictor variables, and for the multicollinearity the degree of correlation between two or more predictor variables were analyzed.

In order to perform estimations and predictions for 222Rn and 222Rn progeny concentrations, the meteorological parameters have been used as independent variables in the multiple linear regression model [128], [129]: air temperature, relative humidity, atmospheric pressure, precipitation and wind speed.

The regression model performances for the estimation of the time series values are quantified by the following regression statistics: multiple correlation coefficient (R), coefficient of determination (R2), p-value (significance level), F (test), sum squared residuals (SSR). In case of prediction, the model performance was quantified by means of correlation coefficient R, p-value and SSR [130], [131].

Processed data regarding the radon and thoron progeny concentrations were measured in the year 1996 by the Environmental Radioactivity Monitoring Station Botoșani (ERMS), Romania.

Meteorological data were measured in the same area. The choice of this year is justified by the fact that does not exist missing data for both radon and thoron progeny concentrations and corresponding meteorological data.

Method of Measurement

The measuring method of radon and thoron progeny activity concentrations is based on the aerosol aspiration on glass fiber filters with high filtration efficiency (96-99%), using high volume aerosol samplers with aspiration head located at 2 meters above the ground.

Aerosol sampling and measurement were performed as described in chapter 9, subchapter 1.

Concentrations of radon and thoron progeny used for this study are daily averages obtained from four aspirations.

Time series of daily data was used to develop multiple linear regression model for estimating and predicting these concentrations.

Results and Discussion

Figure 10.1 presents the variation of the monthly average weather conditions (precipitation, atmospheric pressure, wind speed, relative humidity, air temperature) and concentrations of ^{222}Rn and ^{220}Rn progeny.

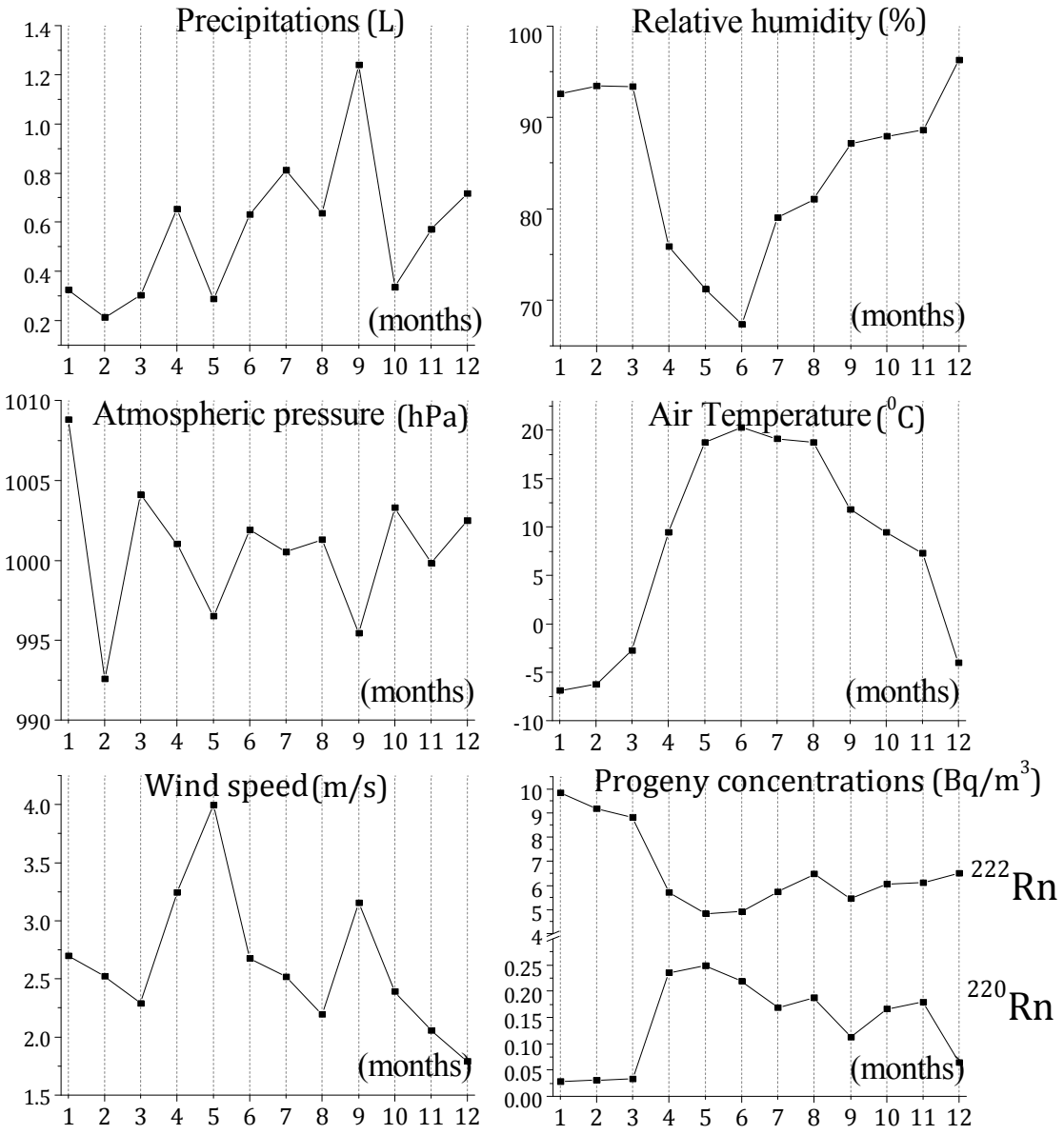

Figure 10.1 - Variation of monthly average concentrations of ^{222}Rn, ^{220}Rn progeny concentrations and meteorological variables

As can be seen from Figure 10.1, both ^{222}Rn and ^{220}Rn progeny are related with meteorological data. In case of ^{222}Rn progeny maximum concentrations were in winter time - January due to less prounounced quantities of precipitations. Also, when were large amounts of precipitations in September, concentration of ^{222}Rn and ^{220}Rn progeny has decreased [118], [128], [129], [132].

The air temperature is the parameter with the greatest influence in the determination of progeny concentrations.

In case of thoron progeny, due to the small half-live (56s), there were no significant changes in concentrations in winter, the rest of concentrations being similar to that of radon.

Multiple linear regression model

In order to find the linear relationship between a dependent variable and several independent (or predictor) variables by using the multiple linear regression model it was studied the performances of estimations and predictions of radon and thoron progeny concentrations considering meteorological data [133], [134].

Multiple linear regression model can be used for estimations and predictions, with statistical significance, only if the independent varriables are not in a relation of collinearity (correlation coeficients close to 0.99 and larger) and if two or more predictor variables are not in a relation of multicollinearity - Variance Inflation Factor (VIF) must be <10 [133], [135], [136].

Multiple linear regression model assumes a linear relationship between the progeny concentration as dependent variable and the meteorological data as predictor variables [128], [133]

$$y(t) = \beta_0 + \beta_1 x_1(t) + \beta_2 x_2(t) ..... + \beta_n x_n(t) + \varepsilon_c \qquad (10.1)$$

where:

$y(t)$	- the value of the dependent variable
$x_1(t), x_2(t), ... x_n(t)$	- the current variable, value of the dependent variable at a time t given by the values of the independent variables at the same moment (the time corresponding to the value of the time series)
β_0	- constant regression
β_n	- predictor coefficient $x_n(t)$
ε_c	- time error

Estimation and Prediction of ^{222}Rn and ^{220}Rn progeny

The estimation of progeny concentrations is made by using the analytical expression of the multiple linear regression with the corresponding meteorological variables from the time period on which the regression is defined.

Multiple linear regression model assumes a linear relationship between the progeny concentration as dependent variable and the meteorological data as predictor variables.

From Figure 10.2, containing representative months for each season, it can be clearly seen, that the estimation values generated by the multiple regression model, are very close to the measured values. This means that the considered predictor variables may describe well the radon and thoron concentration dynamics in the respective months.

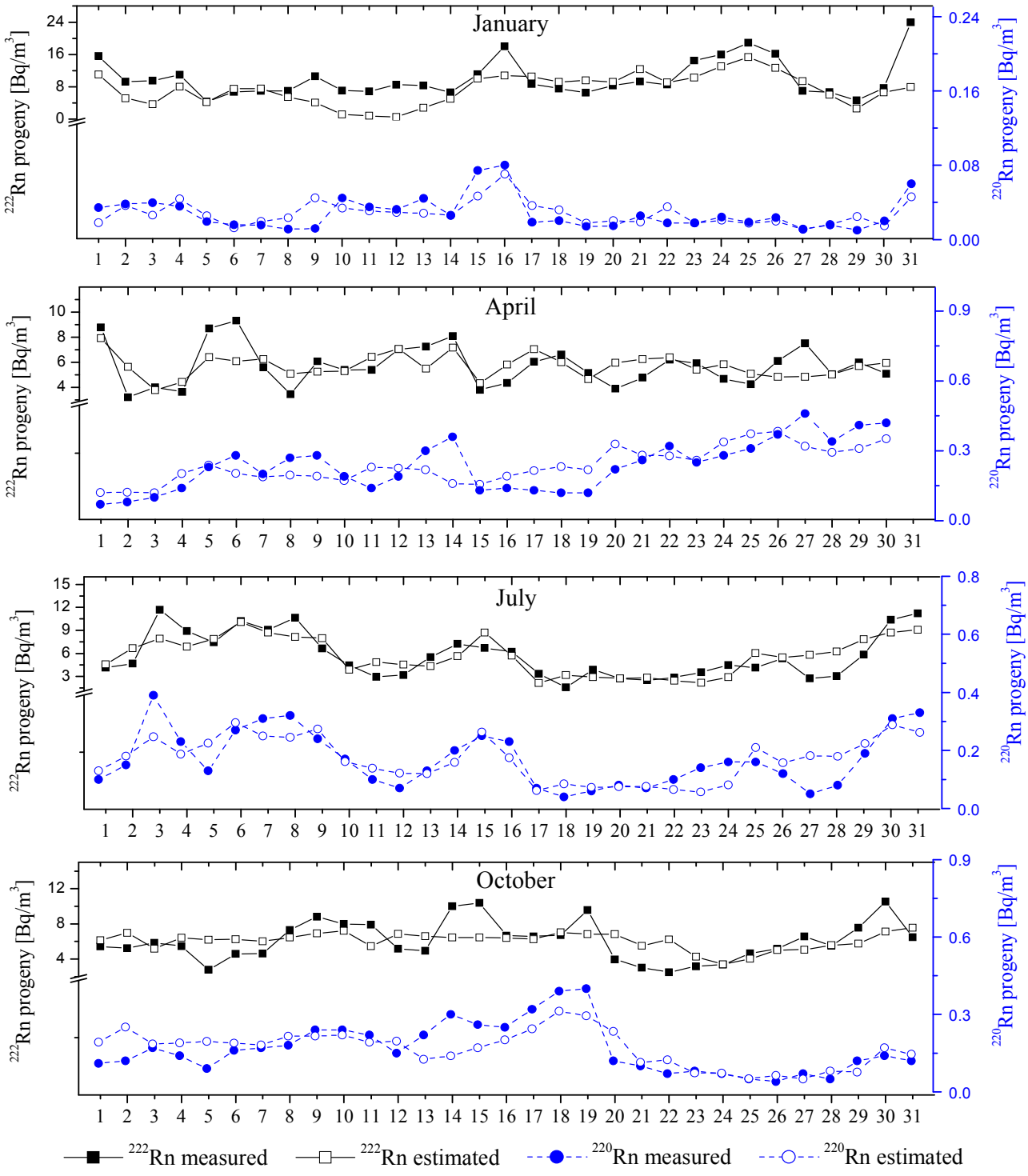

Figure 10.2 - Estimation of concentrations of ^{222}Rn and ^{220}Rn progeny

Making use of the dedicated software [137], [138] and Excel, the regression coefficients are determined by the least square method. In the Table 10.3 the regression statistics for the estimation of the ^{222}Rn and ^{220}Rn progeny concentrations for a representative month of each season, are presented.

Table 10.3 - Statistics used to estimate progeny of ^{222}Rn and ^{220}Rn

Month	Ecuation statistics for ^{222}Rn progeny					Ecuation statistics for ^{220}Rn progeny				
	R-multiple	R^2	p	F	SSR	R-multiple	R^2	p	F	SSR
Ian '96	0,730	0,534	$1,2e^{-3}$	5,730	298,63	0,731	0,534	$1,1e^{-3}$	5,753	0,004
Apr '96	0,559	0,313	$8,9e^{-2}$	2,188	54,139	0,690	0,476	$5,6e^{-3}$	4,375	0,175
Jul '96	0,809	0,655	$3,6e^{-5}$	9,495	89,407	0,781	0,611	$1,4e^{-4}$	7,867	0,107
Oct '96	0,434	0,188	$3,6e^{-1}$	1,162	121,93	0,772	0,521	$1,5e^{-3}$	5,453	0,134

One may notice that the multiple correlation coefficients has the greatest values in July with R values equal to 0.809 with very high level of significance ($p<0.01$) for both radon and thoron progeny concentrations. This proves that the corresponding predictor variables are describing quite well the variability of radon progeny in this month.

Regression statistics in Table 10.3 shows that for estimating concentrations of radon and thoron progeny, the highest values of R-Multiple were obtained in July due to the fact that radon, with comparable life time ventilation of the entire planetary boundary layer is very sensitive to temperature that induces vertical transport of air masses.

The prediction is achieved by using the analytical expression of regression obtained for a previous time period, with predictor variables from the time interval on which the prediction is done [128], [138]. The predictions of the progeny concentrations $[y]_{N+1}$ on the time interval N+1, were performed by using the following analytical expression [128]:

$$[Y]\ N+1 = \beta_0 + \beta_1\ [x_1]\ N+1 + \beta_2\ [x_2]\ N+1 + ... + Bn\ [x_n]\ N+1 \qquad (10.2)$$

$[x_1]\ N+1$, $[x_2]\ N+1$,... $[x_n]\ N+1$ are the values of the independent variables in the time frame N +1.

From the Figure 10.3 where the prediction values generated by the multiple regression model are compared with measured values showed better results obtained for the first part of the representative months it can be clearly seen because of the changing of meteorological conditions from the prediction period considering the fact that the equation constants are calculated considering the meteorological data from the previous month. This means that the considered model for estimation and prediction may describe well the radon and thoron progeny concentrations dynamics in the respective months.

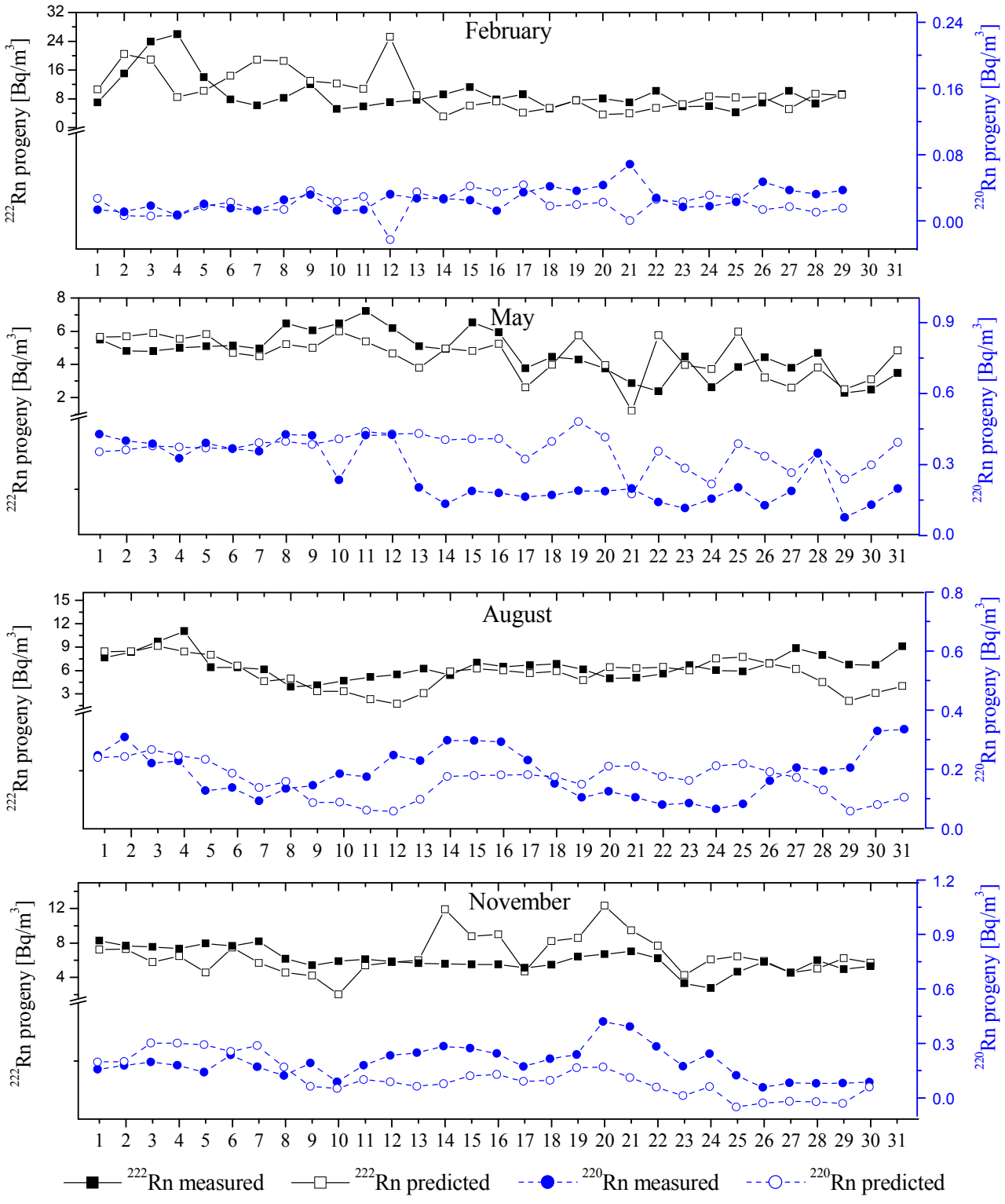

Figure 10.3 - Prediction concentrations of ^{222}Rn and ^{220}Rn progeny

In Chapter 11 was studied the **migration of ^{137}Cs from soil in moss** and soil in different regions of Romania and the radionuclide transfer factor from soil to moss. It was analyzed the spatial and temporal distribution of the samples, correlated with the average monthly amount of precipitation recorded during sampling and the specific activity obtained as a result of gamma spectrometric analyses. Also, it was studied the dependence of the transfer factor of ^{137}Cs from soil to moss, considering the altitude and growth environment of the moss.

The main anthropogenic sources of environmental pollution with artificial radionuclides are as a result of atmospheric deposition after nuclear weapons testing and nuclear power plants accidents. This action introduced among others in the environment, ^{137}Cs with an uneven distribution and transport pathway which is also a common behavior for other radionuclides.

Study under this chapter aimed mainly the analyze of ^{137}Cs radionuclide concentration in mosses and soil in different areas of Romania and the transfer factor of ^{137}Cs from soil to moss.

Systematic analysis of transfer factor of radionuclides from soil to plants is particularly important when assessing the dose with various mathematical models.

Materials and Methods

Sampling of soil and vegetation was done in August-September 2009. The choice of sampling period is important because the radionuclide activity depends on various factors, including weather conditions [142]. Moss and soil under moss samples were taken from unexposed areas of industrial pollution and traffic, as far away from urban areas.

Measurements were done by usding a high purity and resolution broad germanium gamma spectrometer (HPGe), from Canberra, model BE 3820, n-type, planar, covering a range from 3 keV to 3MeV with nominal efficiency of 35%. Energetic resolution at 5.9 keV was 450 eV, at 22 keV of 750 eV and at 1332.5 keV of 2100 keV.

The Genie 2000 software was used for analysis. Energy calibration was performed using spectrum's peaks and efficincy calibration was performed by Canberra's ISOCS. Efficiency calibration verification was performed using reference materials from IAEA. Corrections for geometry, density, matrix and summation were performed.

Results and Discussion

13 sampling locations were chosen for the study. Considering the specifics of the sampling are, they can by devided in three support mediums: wood, rocks/cliffs and trees respectively.

For the purpose of calculation the transfer factor (TF) of ^{137}Cs from soil to moss, reported results for both samples types was done at dry weight (d.w.).

In the analyzed samples, ^{137}Cs concentration in moss is ranging from 8.019 ± 1.212 Bq/kg d.w., up to 259.823 ± 32.146 Bq/kg d.w., respectively from 20.985 ± 2.811 Bq/ kg d.w. and 899,437 ± 110,387 Bq/kg d.w. for the soil samples beneath the moss. Results are accompanied by expanded uncertainty expressed for a coverage factor of k = 2, corresponding to a confidence level of 95 % [101], [102].

From the results presented it is noted that the growing trend of ^{137}Cs activity in samples is directly influenced by the location of the sampling point in relation with the original contamination.

Due to monitoring the uncultivated soil radioactivity, as a result of atmospheric deposition, one can see that ^{137}Cs concentration decreased significantly in the last 23 years after the contamination due to the accident at Chernobyl NPP, from 1986.

Decreased concentration of ^{137}Cs is shown graphically in Figure 11.3, based on data from the National Environmental Radioactivity Surveillance Network (NERSN) from 1985 to 1993 [37] and 2006-2010 [144].

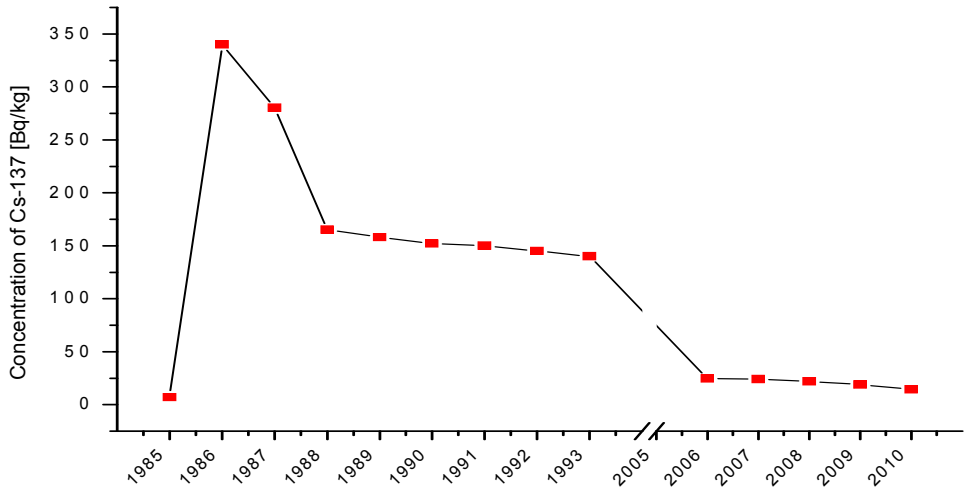

Figure 11.3 - The concentration of ^{137}Cs in soil samples fallow, annual average obtained during 1985 to 1993 [37] and 2006-2010 [144], [145] by NERSN

In figure 11.3 is shown the result of long-term monitoring of ^{137}Cs levels in uncultivated soil samples. The graph shows a pronounced downward trend recorded for this radionuclide, which shows that the only major source of environmental contamination with ^{137}Cs was the Chernobyl accident.

Although the relationship between moss and the soil beneath the moss is in very good as in the previous case, taking into account the sampling altitude, it indicates a medium correlation. Presence of the minus sign in both cases where the sample and the sampling altitude are correlated, indicates that as altitude increases, ^{137}Cs concentration in the samples decreases, which can be noticed in figure 11.5.

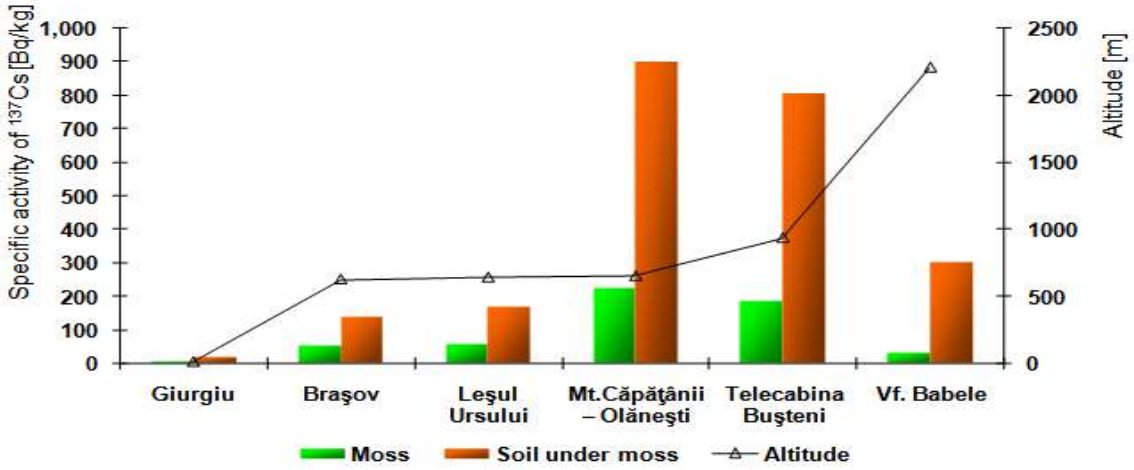

Figure 11.4 - Variation of ^{137}Cs concentration in moss and soil under the moss, sampled from rocks/cliffs

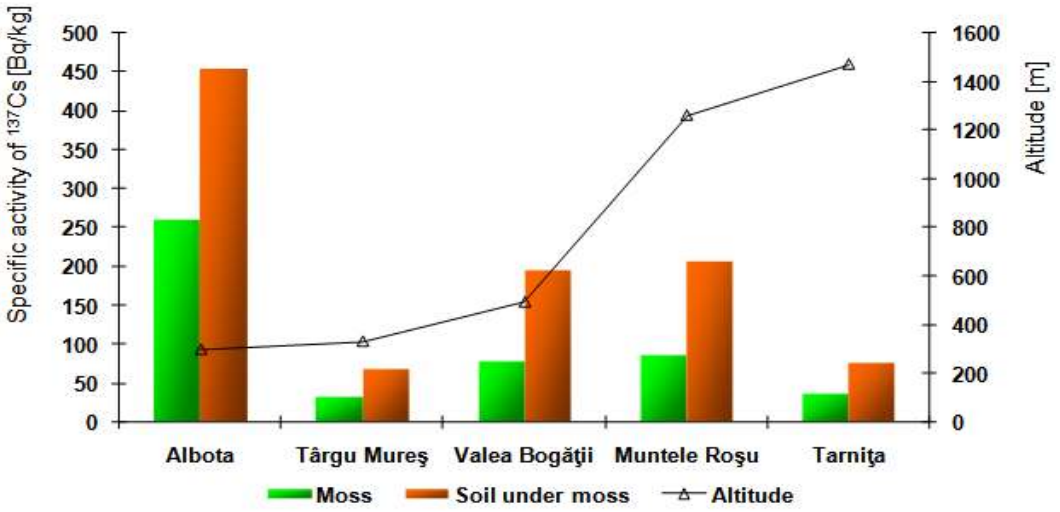

Figure 11.5 - Variation of ^{137}Cs concentration in moss and soil under the moss, sampled from forest areas

In order to assess ^{137}Cs, artificial radionuclide, transfer from soil to moss, the transfer factor (TF) was calculated based on the specific activity of ^{137}Cs in soil and moss samples, taken from the same place. ^{137}Cs transfer factor from soil to moss was calculated using the folowing relation [146], [147], [148]:

$$TF = \frac{\text{radionuclide activity per mass unit of dry vegetation (Bq/kg)}}{\text{radionuclide activity per mass unit of dry soil (Bq/kg)}} \qquad (11.1)$$

The analyzed results for each location are shown in Figure 11.6.

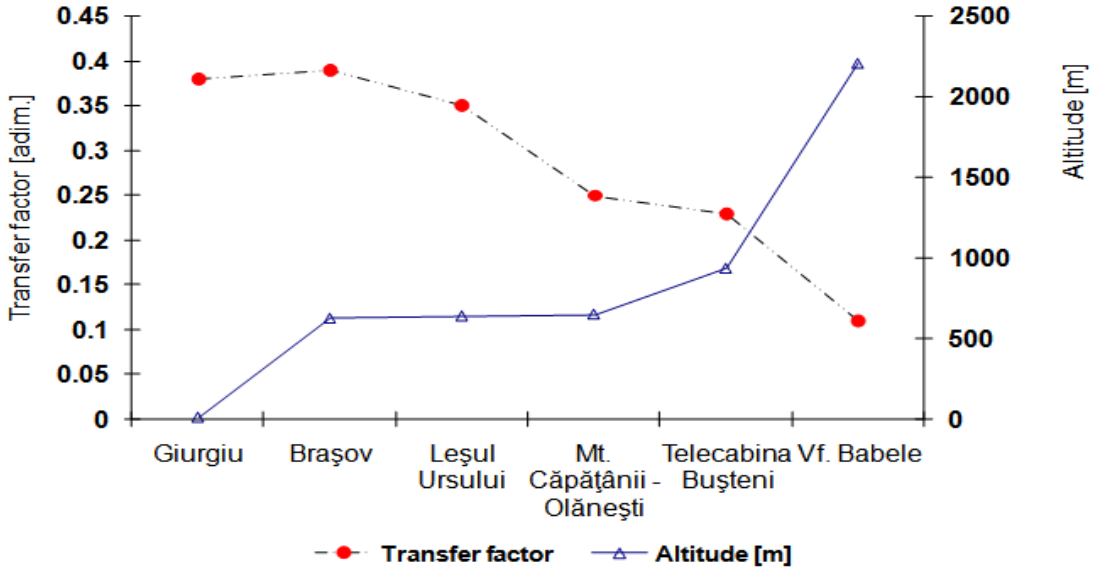

Figure 11.6 - TF of ^{137}Cs from soil to moss depending on the altitude, case of the samples taken from rocks/cliffs

As can be seen from Figure 11.6, the correlation between the TF and the altitude is very good (-0.883) for all sampling points, the minus sign indicating that TF decreases as the altitude increases. This confirms the previous assumption about uncorrelation of the values obtained for samples taken from Babele due to the great difference in altitude.

Conclusions and personal contributions of the thesis are presented in the last part of the paper and highlights the main original results discussed in Chapters 5-11, which were presented in papers published in journals or within national and international scientific events.

Scientific objective of this study was to conduct a survey of the environmental radioactivity (water, air, soil and vegetation) in order to determination the radioactive isotopes concentrations (^{3}H, ^{222}Rn, ^{131}I, ^{137}Cs) using modern devices and for sampling and analysis procedures standardised or original, in some cases.

Conclusions and original contributions are as follows:

In the study on optimizing the preparation of water samples to determine the specific activity of tritium

- Preparation of water samples was studied by two methods. The standardized method takes into account the evaporation of the standard sample in a heating mantles and uses between the flask and condenser a Vigreux distillation column type, leading to a distillation for about 3 hours, while the unstandardized method developed using a rotary evaporator has removed the Vigreux column. The time required in the last case of distillation was about 30 minutes for a sample of 250 mL. Reduction the distilation time due to of the vacuum from the system, which favors the reduction of the boiling temperature of water.
- The study developed in order to validate the proposed method shows that the solution has a high level of confidence, complying with standard parameters and allowing a shorter sample preparation time.
- The proposed method of sample preparation can be extremely useful both for routine analyzes monitoring and emergency situations, when having a result accurately and quickly is crucial.

In the study of tritium dynamics in precipitation

- Given the location near Calarasi and Slobozia from Cernavoda, and their behavior similar for the same weather conditions, it can be concluded that these checkpoints are ideal for monitoring of Cernavoda NPP.
- Although Craiova is more strongly influenced by continental conditions than Slobozia and Calarasi, it follows almost the same trend observed in the other two locations, characterizing atmospheric tritium behaviour virtually in the entire Romanian Plain.
- After data comparative analysis one can see that there is a strong trend of accumulation of tritium in the atmosphere during dry periods and its transfer in the first amounts of precipitation.
- Strong and medium positive correlations were observed in environment during the winter months, leading to the conclusion that tritium transfer in precipitation in the cold season is much slower due to negative temperatures.

In the study to determine the specific activity of tritium in soil

- The case study of this chapter presents an interdisciplinary analysis of soil samples taken from the arboretum of the Institute of Physics and Nuclear Engineering " Horia Hulubei " (IFIN - HH), located about 2 km from the institute and a few kilometers from Bucharest.
- The study presents some experimental data, site-specific, related to the occurrence of tritium in an area subject to the influence of different nuclear activities including routine emission and effluent gaseous tritium from IFIN -HH, in these specific circumstances, tritium issued routinely IFIN -HH is a permanent presence in the area, making it a highly convenient medium.
- The experimental results showed that although the concentration of tritium emissions from IFIN-HH is low, it can be highlighted with higher values than those of the same environmental factors taken as a reference, which is why we analyzed the distribution of tritium in three soil profiles in order to reconstitute the accumulation of information "historic" from the area. This case study was a necessary step in the complex task dedicated arboretum landscape restoration.
- The use of tritium as a tracer for environmental investigations were both determining its concentration in two compartments (soil, vegetation) of the area concerned and as well as its distribution in soil profiles, from the surface to the depth of the groundwater level.
- Tritium specific activity associated with the ground water located in pores taken in the three boreholes, mainly arboretum, emphasizes the accumulation of water in the upper layers (muddy clay, gray - blackish, medium fine consistency), approx. 1.5 m depth, demonstrating the surface water contamination rather than of the groundwater.
- High level of concentration of tritium in the range of 1.50-1.70 m depth, is due to " residual memory " of muddy clay, associated with the maximum depth reached by the roots of vegetation allows water to support a top layer placed above the extended ground water, thereby developing tree species horizontal distribution of roots and the collapse of pivoting rooted tree species. Blackish gray muddy clay consistency medium plastic, placed at different depths in the three sampling points within the park, shows swampy land origin and the existence of a chain of ponds spread far in the history of this place in the mid-nineteenth century.

In the study the environmental monitoring during nuclear accident or radiological emergency. Fukushima accident case study.

- The presence of artificial radionuclide 131I in the atmosphere was continuously monitored in Romania between 23.03.2011 - 30.04.2011 by NERSN, as a result it was found that required time for the cloud to reach the territory of our country was of 15 days.
- Stopping of the intensive monitoring was done as a result of stabilization of Fukushima and to obtain more than two weeks of results below limit of detection equipment for both types of samples, thus confirming the statements of the Japanese authorities.
- The concentrations of ^{131}I highlighted by gamma spectrometric analysis were comparable to those reported by other countries [111], [112], [113], [114], for atmospheric aerosols and atmospheric deposition.
- Information provided by gamma spectrometric analysis for total atmospheric deposition indicates that Romania was not always under a cloud, as confirmed by the results obtained from the analysis of atmospheric aerosols.
- Weather conditions, the relatively short half-life time of the artificial radionuclide ^{131}I , the distance from the crash site and the measures taken by the Japanese authorities to reduce the effects of the nuclear accident at Fukushima have worked togheter in order to reduce the ^{131}I

specific activity below the equipments detection limit, both for atmospheric deposition and atmospheric aerosols, in individual and accumulated samples, at different time intervals from one day to a maximum of one month.

- In order to verify the possible effects that the presence of ^{131}I could have on human health, the dose estimation by inhalation have been calcultate taking in consideration the worst case scenario: a person with permanent residence Peak Toaca from Ceahlău Mountain, exposed for one year to the maximum value (3.494 ± 0.247 mBq/m^3). According to the Radiological Safety Norms, NSR - 01 [115], calculations for population revealed a unsignificant contribution of inhalation exposure from ^{131}I, at the sampling points, of over 1000 times smaller than that of the natural background radiation.

In the study of atmospheric radioactivity correlation with meteorological variables

- The experimental results obtained in this study emphasizes the need for ongoing monitoring of environmental radioactivity to determine the natural background level of each type of sample, which is strongly influenced by weather conditions. This type of information is vital for a potential accident / incident nuclear when necessary to provide timely and reliable data on elevations of radioactivity in different environments, increases wich can not be justified by the locat weather conditions and require the presence of artificial radionuclides in the environment.
- Daily monitoring of environmental radioactivity provides an indication of changes occurring in natural radioactivity and real-time alerting for decision makers.
- Dependence of the samples activities on the meteorological variables was given by Pearson correlation coefficient. Basically, in the atmosphere, the correlation coefficient indicating the degree of dependence of the concentration of gaseous substances or solids (as aerosols) of the meteorological variables.
- In terms of air radioactivity, an influence is given by the natural radioactivity of soils influenced by the content of radium or radon in soil, thoron and their descendants.
- Atmospheric natural radioactivity is directly influenced by the weather, in particular air radioactivity, while the soil and vegetation are less affected. There is an increasing total beta radioactivity values in soil and vegetation, when radionuclides are brought by rainfall and wind speed. This means that changes in the level of natural radioactivity can occur only if a nuclear accident or radiological emergency.

In the study estimating and predicting concentrations of ^{222}Rn and ^{220}Rn progeny using weather variables

- In terms of prediction evaluation criteria, there were used SSR and the correlation coefficient (R) between predicted progeny concentrations and measured concentrations, the appropriate level of significance p and relative error for each value of the predicted concentration.
- Performances of presented predictions indicates that both concentrations of radon and thoron progeny are well described by multiple linear regression model, except for radon in November (N = 0,176). The explanation lies in a great variety of weather conditions for the month in which the prediction was carried out - November time period used to determine the predictive equation - October
- Prediction analysis on progeny concentrations showed the limitations of the prediction procedure are muche more pronounced in case of ^{220}Rn progeny concentrations, due to the much lower concentrations than those of ^{222}Rn progeny, as well as the much lower

concentrations in the air. Thoron progeny concentrations are more sensitive to changes in the independent and dependent parameters used for estimation and prediction.

In the study of soil ^{137}Cs migration in moss

In this study we analyzed the distribution of cernobian origin of artificial radionuclide, ^{137}Cs in moss samples and soil samples under moss on rocks / cliffs, forest areas and trees throughout Romania.

Although the radioactive decay phenomenon prevails at all sampling locations there were identified two other antagonic phenomena which occurs in parallel with this: the accumulation, the decrease of ^{137}Cs specific activity in samples, according to their position.

Individual analysis of samples taken from the three environmental media (rocks/cliffs, forest areas and trees) revealed the following:

- ^{137}Cs concentration in the soil under the mosses, grater than in mosses, is the highest for rocks/cliffs and the lowest for trees, which is justified by cesium affinity to strongly adhere on carbonates from rocks and soil.
- in the case of the rock samples there is a good positive correlation between the height and the level of contamination, while the samples taken from the areas of forest, the correlation is negative.
- In the case of transfer factor, the decreasing trend according to altitude is clearly visible for all samples.
- the samples from the trees are different, the soil (most likely accumulated from dry atmospheric deposition) has only about 9% more 137Cs content than in moss. This can be explained by the extremely small amount of carbonates from the existing soil under the moss, which basically allowed plants to balance their levels of cesium in the soil.

The results of the case studies presented in this thesis allows the optimization of the monitoring and interpretation of data on environmental radioactivity (natural and artificial).

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Annex

PUBLISHED PERSONAL CONTRIBUTIONS

Scientific considerations mentioned above are the personal contributions that have been published:

• in ISI journals:

1. Simion, C. A.; Simion, E.; Păunescu, N.; Mocanu, N.; El-Shamali, S.; Burda, Ș.; Manta T.; Ionescu, D., *The use of tritium resulting from nuclear activities as environmental tracer*, Journal of Labelled Compounds & Radiopharmaceuticals, 53 (5-6), pp. 477-480, 2010
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• in collections of papers at international conferences:

1. Simion, E.; Simion, F.; Verbuță D.C.; Leahu, E., *Long-term variation of outdoor radon concetration in Bacău*, IRPA Regional Congress for Central and Eastern Europe, 24-28 September 2007, Brașov, Romania
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• in collections of papers at national conferences:

1. Simion, E.; Simion, F., *Monitorizarea radioactivității factorilor de mediu în zonele cu fondul natural modificat antropic*, PROTCIV 2009, Sesiunea de comunicări științifice cu tema: "Protecția civilă, apărarea împotriva dezastrelor și gestionarea situațiilor", 19 februarie 2009, Snagov, România, Ed. Printech, pp. 370 - 378, ISSN 1843-5343
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