EVOLUTION OF ANTHROPOGENIC RADIONUCLIDES CONCENTRATIONS ON THE SITE OF RATEN ICN PITESTI (ROMANIA) AFTER 32 YEARS FROM THE CHERNOBYL MAJOR NUCLEAR ACCIDENT

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Abstract

The world's electricity is largely supplied by nuclear power plants (NPPs), which brings significant advantages but also raises challenges, the possibility of a severe nuclear emergency being one of the most important drawbacks. This led to a pertinent statement that no nuclear power plant is 100% safe. The accidents at Chernobyl and Fukushima NPPs are good examples demonstrating that despite precautionary measures, there is no zero risk associated with the operation of NPPs. The objective of this study was to present the evolution of anthropogenic radionuclides resulting from a major nuclear accident, specifically the Chernobyl nuclear accident, where the radionuclide of interest was Cs-137. Keeping this objective in mind, soil and vegetation samples were collected, prepared and measured from 5 external sampling points around the site of the Institute for Nuclear Research (RATEN ICN Pitesti, Romania).The evolution of activity concentrations of Cs-137, in both soil and vegetation samples, has been graphically represented for monitoring locations during the period 2018-2023. For the soil samples, Cs-137 concentration average values measured in the Laboratory of Radioprotection, Environmental Protection, Civil Protection and Medical Analysis at RATEN ICN Pitesti were graphed against the national average values of Cs-137 concentration reported by the National Environmental Protection Agency. A comparison against previous reported Cs-137 activities measurements carried out in the period 1991-2013 on the ICN Pitesti site was also performed. For the determination of Cs-137 activities, measurements were conducted by gamma spectroscopy using an HPGe detection equipment. Cs-137 activities in soil and vegetation will continue to be monitored on the ICN-FCN platform and will be periodically reported to the Arges Environmental Protection Agency, to be included in the annual reports of the National Environmental Protection Agency.

Keywords: *Cs-137, Nuclear accident, Activity concentration, Environmental activity, Monitoring*

Introduction

In the context of the utilization of radiation in various applications and technologies, the implementation of radiation protection measures is essential. Apart from the practical considerations concerning radiation safety and its environmental impact, the public concerns regarding these issues must also be taken into account. Further research in the field is therefore required to create a solid scientific basis that supports all aspects of the risks related to radiation exposure.

An essential element of radiation protection is the monitoring of radiation, which provides methods for assessing individual exposure, critical groups or population groups. This involves a series of activities designed to monitor, control and estimate exposure to radiation and radioactive materials. Although a significant part of these tasks focuses on measuring radioactivity, implementing the results obtained from these measurements is just as important.

Prior to August 6, 1945, the date of the first man-made nuclear explosion, artificial radionuclides were absent from the surrounding environment. Subsequent to this date, nuclear testing and accidents at nuclear facilities led to their widespread use in all environmental compartments.

Looking back into the past, we may specify the top five (5) nuclear accidents or major events that have affected the history. The Chernobyl nuclear accident occurred on April 26, 1986, and was classified at the highest level on the INES scale (International Nuclear and Radiological Event Scale), namely at level 7. Similarly, the Fukushima Daiichi disaster, which occurred on March 11, 2011, was also rated at level 7 on the INES scale. Despite sharing the same rating on the INES scale, the triggering causes were completely different. Chernobyl tragedy happened due to a human error whereas a large-scale tsunami combined with an earthquake of 8.9 magnitude on the Richter scale caused the Fukushima Daiichi accident. Another major nuclear accident, the most serious one in the USA, occurred on May 28, 1979, at the Three Mile Island plant, due to partial meltdown of the nuclear reactor core, leading to its explosion. It was rated at level 5 on the INES scale.

The third most powerful nuclear accident in history, rated at level 6 on the INES scale, and occurred on September 29, 1957, at the Mayak nuclear facility in the former USSR, due to a malfunction in the cooling system of a liquid waste tank (70-80 tons). The most catastrophic nuclear accident in British history happened at Windscale on October 10, 1957; it was caused by an explosion, and was rated at level 5 out of 7 on the INES scale [1].

An emergency at a nuclear power plant can lead to the release of a significant amount of radioactive materials. For instance, Chernobyl accident was caused by two thermal explosions that released into the atmosphere a huge amount of radioactive material (fission products, aerosol particles and water vapours, unburned nuclear fuel, neutron activation products) which was transported by the radioactive cloud according to the atmospheric circulation mainly over Ukraine, Belarus, Russia, Central and Eastern Europe, and Scandinavian countries [2]. During first year after the accident, the main contributors to the radioactive exposure were the short and medium half-live radionuclides (as ^{132}Te, ^{132}I, ^{131}I, ^{140}Ba, ^{106}Ru, ^{106}Rh, ^{103}Ru, ^{103}Rh, ^{136}Cs, ^{134}Cs, etc) which overshadowed the long-live ^{137}Cs contribution. It was estimated that half of the quantity of Caesium and Iodine, and 5% of the remaining radioactive elements present in the reactor were released into the atmosphere. Thirty-eight years later, the consequences of the Chernobyl accident are still being discussed, despite the significant gradually decreasing of the contamination with ^{137}Cs [2, 3].

The behaviour of radionuclides that are accidentally released into the environment is influenced by a number of factors, including the terrain and its utilization, precipitation, and yearly average air temperature. There are significant differences between the climate and geography of the contaminated areas at Chernobyl and Fukushima. For instance, the annual precipitation for Fukushima is higher than the one corresponding to Chernobyl (about twice as much). These differences led to a faster migration of radio-caesium in the Fukushima area compared to the Chernobyl area. In Chernobyl, the catchment areas are characterized by flat terrain and minor slopes, whereas in Fukushima, they are characterized by steep and hilly terrain. One notable aspect of the Chernobyl disaster was the radioactive contamination that resulted from fuel particles being released into the environment. The concentration of fuel particles decreased with increasing distance from the reactor [1].

The discharge of extremely hot and radioactive fuel particles into the environment was the main distinguishing characteristic of the radioactive contamination that followed the Chernobyl accident. These particles were formed due to the shockwave generated by the explosion, temperature gradient and fuel oxidation. The fuel particles deposited as a result of the Chernobyl accident ranged from hundreds of microns to a fraction of a micron. These particles have far less activity than fuel particles [4].

The EU countries have to perform the environmental radioactivity monitoring and to check the levels of radioactivity in air, water, soil, and food, according to the Euratom Treaty, article 35.

Monitoring might be performed continuously or dependent on environmental sampling and laboratory analysis. Typically, there is a national monitoring programme that covers the entire country, as well as site-specific programs developed for monitoring around nuclear power plants and other nuclear facilities (such as uranium enrichment plants, nuclear fuel factories, spent fuel reprocessing installations, spent fuel storage facilities, etc). The mobile monitoring in also considered, in case of a radiological emergency [5].

The European Commission keeps a database of monitoring organizations and facilities and verifies the monitoring arrangements. Additionally, the database includes details on areas in EU member states that are radioactively contaminated [6]. The Radioactivity Environmental Monitoring database (REMdb) was established in 1988 to collect and archive environmental radioactivity data generated post Chernobyl disaster. This database has two main objectives, as follows: i) to preserve historical records of the Chernobyl major nuclear accidents for future scientific research, as well as ii) to store the EU states' data on radioactivity monitoring for environmental reports (contain information on radioactivity levels in the European Union's environment, according to Euratom Treaty, articles 35 and 36) [6]. The Laboratory of Radioprotection, Environmental Protection, Civil Protection and Medical Analysis at RATEN ICN Pitesti (Romania) is part of the monitoring organizations included in REMdb, participating in interlaboratory comparisons regarding environmental radioactivity.

After the Chernobyl nuclear accident and migration of the radioactive cloud, most of the Romania's surface was contaminated with Cs-137 (national average values below 2 kBq /m^2), Southern part of the country being the most contaminated. There were large areas in Oltenia, Banat, North-eastern Moldova, Central and Northern Muntenia where contamination was higher than the national average, up to 10 kBq/ m^2; in some locations the contamination exceeded 10 kBq/ m^2 at ground level [7]. The atmospheric radioactive deposition values were more three orders of magnitude higher compared to the values registered before the accident. Cs-137 concentration in untilled soil samples ranged from 350 Bq/kg to over 1000 Bq/kg in the Northern Gorj County. The highest Cs-137 concentrations were detected at Toaca-Ceahlau Mountains, Targu Mures, Gheorghieni and Parang, reaching a maximum of 2500 Bq/kg dry soil [7]. I-131, Cs-137, Cs-134 and Sr-90 from aerosols and deposition rapidly decayed both in plants and animals; Sr-90 was less than 10 Bq/kg, while Cs-137 and Cs-134, and I-131 have values of several hundred Bq/kg and nearly 1 kBq/kg, respectively [7]. Many years after the accident, Cs-137, Cs-134 and Sr-90 contents in food were still detectable (1 Bq/kg in the period 1990-2003) [7].

In terms of soil contamination, the highest Cs-137 and Cs-134 deposits were measured in Romania on the direction Northeast-Southwest direction, with some values exceeding 80 kBq /m2, as previously reported in [2]. The direction mentioned in [2] for the passage of the radioactive cloud over Romanian territory was confirmed by measurements taken in the south of Italy, Greece, Albania and former Yugoslavia.

The distribution of Cs-137 throughout Romania after the Chernobyl accident was investigated in 2016 by using high resolution gamma ray spectrometry in the low background Slănic-Prahova laboratory on about 750 soil samples collected from the Romanian territory [3], and the results were compared with data collected in 1993 on 62 soil samples. From 43 ± 2 TBq for 1993 determinations to 14.1 ± 0.7 TBq for 2016 determinations, the total Cs-137 inventory integrated over the Romanian territory decreased by a factor of 3 ± 0.3. The 2016 determinations of Cs-137 inventory reached a maximum of 94.2 kBq /m^2, in comparison to 1993 values revealing a maximum of 187 kBq/ m^2 [3].

Cs-137 concentrations after the Chernobyl accident was previously reported for the period 1991-2013 based on soil, vegetation and atmospheric deposition samples collected from eleven (11) monitoring locations distributed on 10 km distance around the institute site [8]. The samples were analysed by gamma ray spectrometry and the results revealed significant variations of the Cs-137 activity concentration ranging from 190 Bq/kg dry to 325 Bq/kg dry (soil samples) and from 2 Bq/kg dry to 17 Bq/kg dry (vegetation samples), respectively [8].

In present paper, the evolution of the Chernobyl originated Cs-137 activity concentration in the period 2018 - 2023 was studied and compared to national average values reported by the NEPA. Present data were also compared against the previously reported ones for the period 1991-2013. The soil and vegetation samples were collected from monitoring locations distributed on the site of the RATEN ICN Pitesti, their analysis being performed by high resolution gamma ray spectroscopy with HPGe detector.

MATERIALS AND METHODS

Nuclear accidents have led to significant increases in the concentrations of Cs-137 in certain uncultivated soil samples. In Romania, these elevated levels have been observed in regions where heavier precipitation occurred during the Chernobyl accident. Concentrations of other activation and fission products (such as C-14, S-90, Cs-134 and Pu-239 – mentioning only those with medium to long half-lives) generally exhibit lower values than recorded Cs-137 [9].

The terrestrial radioactivity is caused by the interaction between the isotopes present within the composition of the Earth's crust. Understanding the factors influencing soil radioactivity is of utmost importance. Direct deposition, irrigation with contaminated water, resuspension and seasonal vegetation cycles are processes through which soil can become contaminated.

In the context of soil sampling, the selection of sampling points is governed by considerations pertaining to the natural terrain type. These points are carefully chosen to avoid areas susceptible to flooding or surface disturbances. Ideally, sampling locations are situated within arable land. They should be at least 50 meters away from significant vegetation (large trees), buildings, houses or industrial facilities, which may transfer environmental contaminants/ pollutants, thereby impacting soil integrity.

Thus, for soil sampling, five sampling points were selected, arranged within a radius of 10 km around the RATEN ICN Pitesti site [10-12], as presented in Table 1. The concentration of Cs-137 was determined from the surface layer of soil, up to a maximum depth of 30 cm. In order to obtain representative soil samples, it is imperative to collect about 1 kg of soil from areas that have not been artificially contaminated or subjected to agricultural processing. When the sampling area is covered with vegetation, it is necessary to remove the vegetation along with a 1 cm layer of soil, before collecting the sample. In addition, the roots must be removed through sieving if they could not be initially extracted by pulling. Where there is no vegetation covering the soil, a 1cm layer of soil is removed before the sample is taken. When dealing with soil covered with snow or frozen, the sample should be collected only after both the snow and the ice have been removed [13].

Cs-137 activity concentrations evolution in soil and vegetation samples collected from the locations mentioned above was previously reported by the authors in the study [8], for the period up to 2013.

Table 1. Locations for Cs-137 activity concentration monitoring in soil and vegetation [10-12]

Nr. crt.	Investigation point	Location	Sampled medium	Frequency
1.	SVE1	Mioveni-in the area of the houses on Bugeag street	soil, vegetation	yearly
2.	SVE2	NE extremity of the DACIA platform	soil, vegetation	yearly
3.	SVE3	Next to the ICN-Mioveni road at a distance of 1150 m from the Reactor chimney	soil, vegetation	yearly
4.	SVE4	Next to the ICN-Treatment Station road at a distance of 1150 m from the Reactor chimney	soil, vegetation	yearly
5.	SVE5	Near ICN-Treatment Station road at 100 m upstream from the Station	soil, vegetation	yearly

Once collected, the samples must be processed in the laboratory. Any remaining vegetation or stones are removed, and the resulting sample is weighed. Subsequently, it is crushed and dried in an oven at a temperature of 110 °C until it reaches a constant weight.

Once dried, the sample is moistened and sieved through a fine mesh. In this manner, the sample is prepared for measurement via gamma spectrometry to determine the concentration of Cs-137.

For measurement purposes, the samples are placed in boxes (cylindrical boxes with a diameter of 7.4 cm, made of polyethylene), which are each labelled with the following information: sample type, date of sampling, sampling location, mass of the collected sample, mass of the dried sample and mass of the sample in the box [13]. The methodology for sampling, processing, and measurement is according to the internal procedures of the Laboratory of Radioprotection, Environmental Protection, Civil Protection and Medical Analysis at RATEN ICN Pitesti.

The measurements were performed using an HPGe detection equipment with an efficiency of 25%. Calibration of the equipment was conducted using calibration standards which contain gamma-emitting radionuclides ranging from 59.57 keV to 1332 keV (Am-241, Co-60, Cs-137, Be-133). Considering the varying densities of the soil samples, adjustments to the efficiency were required. These adjustments were performed using the EFFTRAN software.

Due to the low radioactivity levels of the samples, the chosen measurement geometry entails placing the source in direct contact with the detector.

For the environmental monitoring of a nuclear facility's surroundings, it is imperative to ascertain the concentrations of artificial radionuclides. Among these, the radionuclide of interest is Cs-137, resulting from the Chernobyl accident. Since vegetation constitutes a direct source for humans through the food chain, understanding its contamination mechanism is extremely important. The type of vegetation and the species of plants are important, as the behaviour of radioactive materials depends on these factors. The maximum capture of particles deposited on vegetation occurs through the inflorescence of plants.

Radionuclides enter vegetation through two mechanisms: direct deposition into the atmosphere and root absorption by plants of radionuclides from soil. The type of vegetation, topography and its height are factors influencing the retention of radioactive products by spontaneous vegetation. Equally important is the soil type, due to the root absorption mechanism, as roots adhere differently in various soil types.

Similar to soil sampling procedures, vegetation samples are collected from identical sampling locations and within the same timeframe. About 1 kg of spontaneous vegetation is collected, keeping only the aerial parts, which must be free of debris such as stones or sand. When it comes to cultivated vegetation, only the relevant parts are taken out, and underground portions are carefully washed to remove any remaining soil residues. Vegetation samples are placed in boxes and labelled, noting the following information: type of sample, date of sampling, sampling location, mass of the collected sample, mass of the dried sample and mass of the sample in the box [13]. The vegetation approach has also been carried out in compliance with the RATEN ICN laboratory's internal procedures, just like the methods for soil sampling, processing, and measurement.

RESULTS AND DISCUSSION

Soil samples measurements

Figs. 1a) - e) depict the Cs-137 activity concentrations variation in uncultivated soil samples for the considered monitoring locations, within the period of interest for the study, 2018-2023 [14-19].

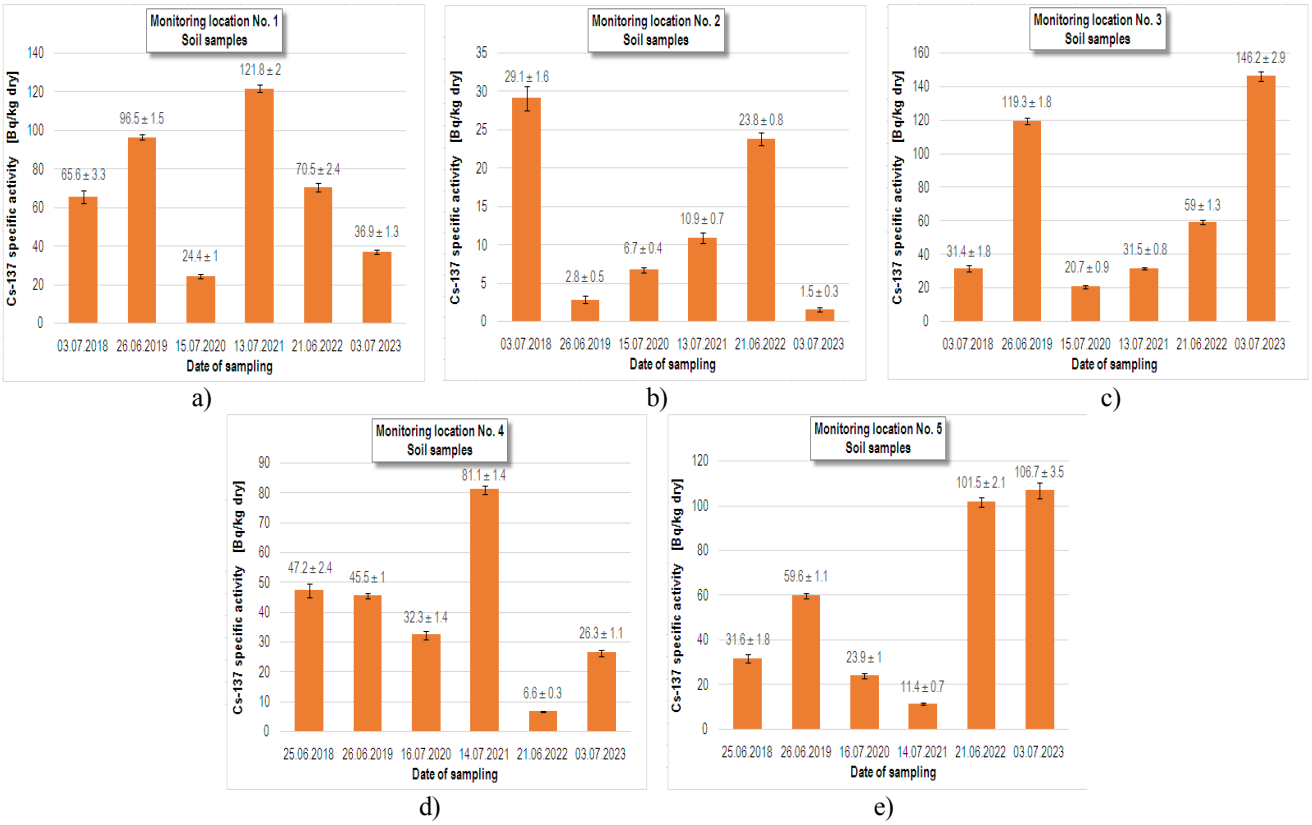


Fig. 1. Variation of Cs-137 activity concentration [Bq/kg] in soil samples from monitoring points:
a) SVE1; b) SVE2; c) SVE3; d) SVE4; e) SVE5

The highest value of Cs-137 activity concentration in the soil samples was registered in 2023 - monitoring location SVE3 (146.2±2.9 Bq/kg). Other high values (over 100 Bq/kg) were measured in 2021 - monitoring location SVE1 (121.8 ±2 Bq/kg), in 2019 - monitoring location SVE3 (119.3± 1.8 Bq/kg), and for the monitoring location SVE5 in 2021 (101.5±2.1 Bq/kg) and in 2023 (106.7±3.5 Bq/kg), respectively.

The lowest values for Cs-137 activity concentration were in the range (1.5±0.1 - 29.1±1.6) Bq/kg, being measured in soil samples collected from the monitoring location SVE2.

Considering the location of the selected monitoring points given in Table 1, both the highest and very low values were expected.

A comparison of the Cs-137 activity concentrations measured in soil samples in the period 2018-2023 for the selected monitoring points was performed and the highest and lowest values measured are presented in Table 2.

The highest Cs-137 activity concentration in soil samples on the RATEN ICN Pitesti site in the period 2018-2023 was 146.2±2.9 Bq/kg, lower by 55% comparatively to 325.3± 1.8 Bq/kg, the highest Cs-137 activity concentration in soil samples previously reported for 1991 - 2013 period [8].

Table 2. Comparison of Cs-137 activity concentration monitoring in soil for different locations

Monitoring point	Highest Cs-137 activity concentration* [Bq/kg]	Lowest Cs-137 activity concentration* [Bq/kg]
SVE1	121.8± 2$^{(2021)}$	24.4±1 $^{(2020)}$
SVE2	29.1± 1.6$^{(2018)}$	1.5±0.3 $^{(2023)}$
SVE3	146.2± 2.9$^{(2023)}$	20.7±0.9$^{(2020)}$
SVE4	81.1± 1.4$^{(2021)}$	6.6±0.3$^{(2022)}$
SVE5	106.7± 3.5$^{(2023)}$	11.4±0.7$^{(2021)}$

* (year) when Cs-137 activity concentration was measured

Vegetation samples measurements

Figs. 2a) - e) depict the Cs-137 activity concentrations variation in vegetation samples collected from the considered monitoring locations, within the 2018-2023 period [14-19].

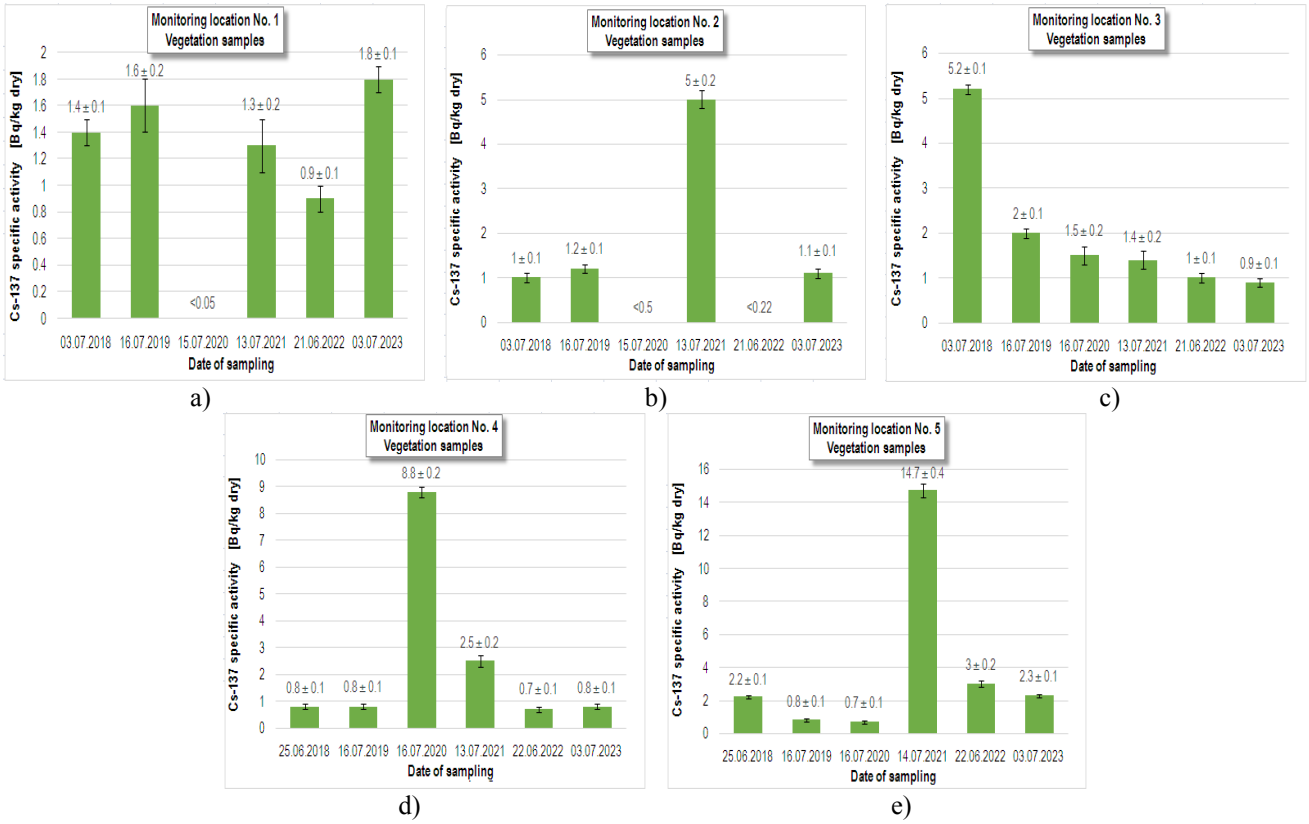


Fig. 2. Variation of Cs-137 activity concentration [Bq/kg] in vegetation samples from monitoring points: a) SVE1; b) SVE2; c) SVE3; d) SVE4; e) SVE5

The highest value of Cs-137 activity concentration in the vegetation samples was registered in 2021 - monitoring location SVE5 (14.7± 0.4 Bq/kg). Another high value (around 10Bq/kg) was measured in 2020 - monitoring location SVE4 (8.8± 0.2 Bq/kg).

The lowest values for Cs-137 activity concentration were in the range (0.1-5± 0.1) Bq/kg, being measured in the vegetation samples collected from the monitoring location SVE2.

Both the highest and very low values were expected as we take into consideration the location of the selected monitoring points on the platform ICN-FCN (see Table 1).

A comparison of the Cs-137 activity concentrations measured in vegetation samples in the period 2018-2023 for the selected monitoring points was performed and the highest and lowest values measured are presented in Table 3.

The highest Cs-137 activity concentration in vegetation samples on the site of RATEN ICN Pitesti in 2018-2023 period was 14.7±2.9 Bq/kg, lower by 14% comparatively to 17.1±3.3 Bq/kg, the highest Cs-137 activity concentration in vegetation samples previously reported for 1991 - 2013 [8].

Table 3. Comparison of Cs-137 activity concentration monitoring in vegetation for different locations

Monitoring point	Highest Cs-137 activity concentration* [Bq/kg]	Lowest Cs-137 activity concentration* [Bq/kg]
SVE1	$1.8\pm0.1^{(2023)}$	$0.5^{(2020)}$
SVE2	$5.0\pm0.2^{(2021)}$	$0.22^{(2022)}$
SVE3	$5.2\pm0.1^{(2018)}$	$0.9\pm0.1^{(2023)}$
SVE4	$8.8\pm0.2^{(2020)}$	$0.7\pm0.1^{(2022)}$
SVE5	$14.7\pm0.4^{(2021)}$	$0.7\pm0.1^{(2020)}$

* (year) when Cs-137 activity concentration was measured

Based on the data published by the Ministry of Environment, Waters and Forests in collaboration with the National Environmental Protection Agency (NEPA) in a report focused on aspects of the environment radioactivity [20] and the national indicators yearly reported by NEPA [14-19], the graph presented below has been developed.

The chart from Fig. 3 shows the evolution of the average values of Cs-137 concentration in soil samples, calculated by applying the decay correction, which takes into account the time elapsed since the accident and the half-life of Cs. These values were around 350 Bq/kg, and the reference date for this value is the year of the accident [20].

The graph also includes the Cs-137 activity concentration values for the soil samples measured at RATEN ICN within the range 2018-2023 [14-19].

Analysing comparatively the evolution of Cs-137 activity concentration in soil samples measured on the platform ICN-FCN against those reported by ANPM for the period of interest (2018-2023), one can notice that the national reported Cs-137 activity concentrations were constantly higher (more than 20%) in comparison with the measured ones, except for the year 2023 when the measured Cs-137 activity concentration was just 2% lower than the national averaged value (149.2 Bq/kg).

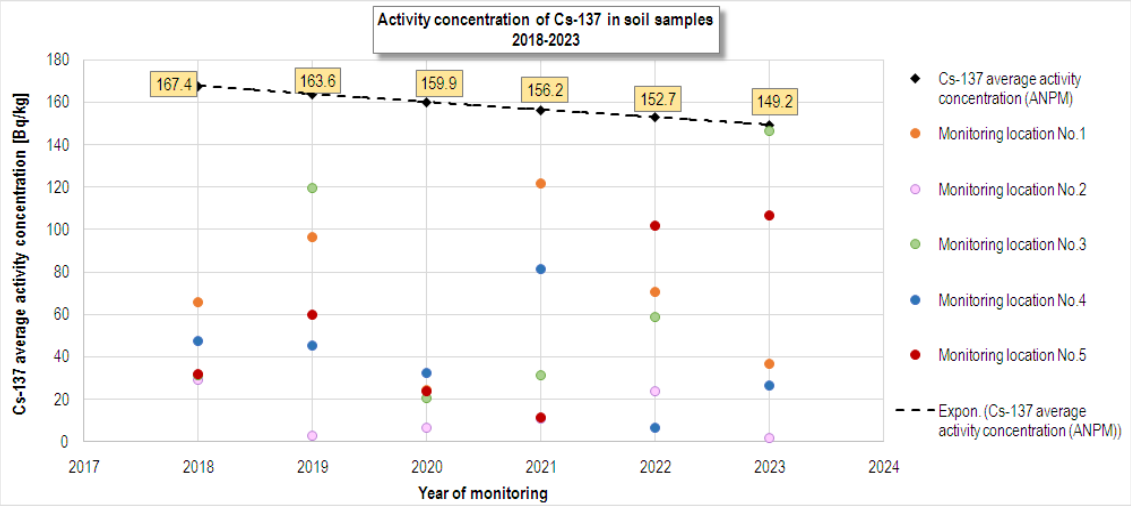


Fig. 3 Evolution of Cs-137 activity concentration [Bq/kg] in soil samples, 2018-2023, [14-20]

Conclusions

Present study aims to report the variation of Cs-137 activity concentration between 2018 and 2023 on the platform of the Institute for Nuclear Research (RATEN ICN) Pitesti, Romania, in order to reveal Cs-137 environmental dynamics following the Chernobyl nuclear accident (1986). To perform the investigation, soil and vegetation samples were collected from the monitoring locations existing on the ICN-FCN platform. The measured Cs-137 activity concentrations were reported according to the monitoring location and period, and compared with the corresponding national average values reported yearly by the National Environmental Protection Agency.

The deposition, condensation and dynamics of displaced material transport influence the behaviour of radionuclides. The parameters determining major variations in radionuclides contaminating soil or vegetation include: humidity, atmospheric turbulence and the chemical form of the isotopes. It is imperative to consider both the soil and vegetation type, as they significantly influence the behaviour of radioactive materials.

From the graphical representations provided, significant fluctuations in the activity concentration of the radionuclide of interest in the study, Cs-137, are evident, visible both in soil samples and vegetation samples. One possible explanation for these fluctuations could be that soil, following certain disturbances generated by the nature, may displace material and reach considerable distances from the point of origin.

The Cs-137 activity concentrations measured in the soil samples are more than10 times higher comparatively to those measured in the vegetation samples collected from selected monitoring points on ICN-FCN Pitesti platform.

The highest value measured in the soil samples was registered in 2023 (146.2 ± 2.9 Bq/kg), for the monitoring point located next to the ICN - Mioveni road at a distance of 1150 m from the Reactor chimney, and is only 2% lower than the national average value reported by the National Environmental Protection Agency. Generally, the measured Cs-137 activity concentration values were much lower than the national averaged values for the period of interest.

As regarding the Cs-137 activity concentrations in the vegetation samples collected from the selected monitoring points on the platform ICN-FCN, the highest value was registered in 2021 (14.7 ± 0.4 Bq/kg), for the monitoring point located near ICN-Treatment Station road at 100 m upstream from the Station.

Considering the extended half-life of Cs-137 (about 30.08 years) and the repercussions stemming from the Chernobyl incident, investigating its environmental dynamics remains a perpetual concern for future endeavours. The monitoring of Cs-137 activity concentration in soil and vegetation will continue on the ICN-FCN platform and the Laboratory of Radioprotection, Environmental Protection, Civil Protection and Medical Analysis at RATEN ICN Pitesti will periodically report the measured values to the Arges Environmental Protection Agency, to be included in the annual reports of the National Environmental Protection Agency.

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