Radiological monitoring of the soil in the proximity of National Radioactive Waste Repository in Różan in years 2014–2023

Radiologiczny monitoring gleby w otoczeniu Krajowego Składowiska Odpadów Promieniotwórczych w Różanie w latach 2014-2023

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Summary: Although Poland does not operate any nuclear power plants, there are two facilities connected to the processing or storing of radioactive substances, which must be under radiological supervision and control. These facilities are the National Centre for Nuclear Research (NCBJ) in Świerk and the National Radioactive Waste Repository (KSOP) in Różan Both of these facilities are monitored by independent laboratories. The concentrations of radionuclides in soil, vegetation, aerosols from the ground layer of air, groundwater and springwater are all assessed in those studies. For a long time, such research was conducted by CLOR. This work compiles the results of such studies from the last 10 years.

Thanks to constant radiological monitoring of the proximity of the KSOP it is possible to determine, whether nuclear waste stockpiled there has an effect on the immediate environment.

This work focuses on the analysis of the radiological measurements of soil samples from years 2014–2023 by comparing the concentration of the natural radionuclides, such as ^{226}Ra, ^{228}Ac and ^{40}K to mean concentration in Poland and Mazowieckie Voivodeship in which KSOP is situated.

Additionally, the deposition of ^{137}Cs, which is an artificial radionuclide left in soil as an aftermath of the Chernobyl Nuclear Power Plant accident, is also measured.

Keywords: Radioactivity of soil, caesium ^{137}Cs, radium ^{226}Ra, potassium ^{40}K.

Streszczenie: *W Polsce pomimo braku elektrowni jądrowych istnieją dwa miejsca związane z przetwarzaniem lub przechowywaniem substancji promieniotwórczych, które podlegają nadzorowi oraz kontroli radiologicznej w otoczeniu tu wymienionych ośrodków, tj.: Narodowego Centrum Badań Jądrowych (NCBJ) w Świerku oraz Krajowego Składowiska Odpadów Promieniotwórczych (KSOP) w Różanie. Obydwa te ośrodki są monitorowane przez niezależne laboratoria. Badane są m.in. gleba, roślinność, aerozole z przyziemnej warstwy powietrza atmosferycznego oraz wody źródlane i gruntowe. Przez długi okres takie badania prowadziło Centralne Laboratorium Ochrony Radiologicznej (CLOR). W pracy zamieszczono wyniki badań gleby z ostatnich 10 lat.*

Dzięki stałemu monitoringowi radiologicznemu otoczenia KSOP można określić, czy składowane tam odpady wpływają na skażenie promieniotwórcze otoczenia.

Praca skupiła się na analizie badań gleby z lat 2014–2023 poprzez porównanie stężeń naturalnych izotopów promieniotwórczych, takich jak: rad ^{226}Ra, aktyn ^{228}Ac oraz potas ^{40}K, do średnich stężeń w Polsce oraz w woj. mazowieckim, w którym KSOP jest umiejscowiony.

Dodatkowo, zbadana została depozycja cezu ^{137}Cs, który jest sztucznym izotopem promieniotwórczym pozostałym jeszcze z czasów awarii elektrowni jądrowej w Czarnobylu.

Słowa kluczowe: *Promieniotwórczość gleby, cez ^{137}Cs, rad ^{226}Ra, potas ^{40}K.*

Introduction

The National Radioactive Waste Repository (KSOP) in Różan is the only facility in Poland designed for the disposal of short-lived low- and intermediate-level radioactive waste, as well as for the interim storage of long-lived waste. In addition, short-lived low-level or intermediate-level disused sealed radioactive sources are also disposed at KSOP.

The primary barrier preventing the release of waste into the environment, e.g. into groundwater and soil, is a system of concrete layers separating the waste from its surroundings. Short-lived waste is placed in metal drums, which are subsequently arranged in designated storage places and backfilled with concrete. Additional protection against precipitation is provided to prevent water from penetrating the concrete, which could otherwise cause corrosion of the structures and lead to the migration of radionuclides into the soil.

According to the applicable Polish and European legal regulations on radioactive waste storage, nuclear facilities such as the National Radioactive Waste Repository must be subject to radiation monitoring. This monitoring covers both the repository site itself and its immediate vicinity.

On request of the Polish National Atomic Energy Agency, the Central Laboratory for Radiological Protection (CLOR) carries out comprehensive measurements aimed at assessing the radiation situation around KSOP. These measurements include samples of soil, grass, aerosols from the near-ground atmospheric layer, as well as spring and groundwater. This paper presents the results of soil measurements conducted in the surroundings of KSOP in the years 2014–2023.

Soil analysis

As part of the radiological assessment of the environment around KSOP, soil samples were collected. Sampling locations were selected in such a way as to allow assessment of the repository's impact on the environment from all sides (Fig. 1) Sampling took place twice a year, resulting in a total of ten soil samples annually. In the samples, radioactive gamma isotopes of natural origin were recorded, i.e. from two natural radioactive decay series and potassium ^{40}K, as well as artificial radioactive gamma isotopes. Among the artificial radionuclides, caesium ^{137}Cs was mainly detected, and in one year the presence of americium ^{241}Am and cerium ^{141}Ce was also observed.

Fig. 1. Soil sampling points in the vicinity of the KSOP in Różan (source: http://www.google.pl/maps).

Sampling procedure

Soil samples were collected in accordance with Fig. 1 from the topsoil layer (10 cm) using metal samplers. One of the seven subsamples (from one sampler) was located at the centre of a circle with a radius of 1 m, while the remaining six subsamples were taken along the circumference of that circle. The collected subsamples were then mixed in a bag and properly labelled. This sampling method is recommended by the International Atomic Energy Agency (IAEA) in Vienna [1].

Sample preparation

Before the measurements were carried out, the collected samples had to be properly prepared. In the first step, the samples were crushed and cleaned of impurities. The prepared soil was then dried in an oven at 105°C for 16 hours, after which it was cooled to room temperature. After determining the total mass of the sample and further crushing, they were sieved through a 2 mm sieve and poured into 0.5 dm^3 Marinelli beakers.

Measurement procedure

The measurements were performed using gamma ray spectrometry with a HPGe semiconductor detector of approximately 40% relative efficiency, operated with GENIE-2000 software. The counting time for each sample was 80,000 seconds, at which the minimum detectable activity in soil samples was 0.05 Bq/kg for caesium ^{137}Cs and 1 Bq/kg for potassium ^{40}K, radium ^{226}Ra and actinium ^{228}Ac.

Measurement results

The gamma radiation spectrum of each sample was analysed using the GENIE-2000 software, taking into account both the efficiency calibration and the previously measured background.

Tables 1–3 present the activity concentrations of natural radionuclides ^{226}Ra, ^{228}Ac and ^{40}K together with associated uncertainties, as well as the mean values of these concentrations for the five sampling points in the years 2014–2023, expressed in Bq/kg.

Table 1. Activity concentration of ^{226}Ra in soil samples measured in 2014–2023 expressed in Bq/kg.

Radium ^{226}Ra	D-1	D-2	D-3	D-4	D-5	Collection date
2014	18.1±0.7 20.4±0.7	14.9±0.6 18.3±0.7	10.6±0.4 11.9±0.5	13±0.4 14.6±06	15.9±0.6 16.1±0.6	21-05-2014 09-09-2014
2015	21.9±0.8 33.4±1.3	31.0±0.9 30.6±1.2	28.5±0.8 22.9±1.0	17.0±0.7 27.6±0.9	38.6±1.0 30.2±0.9	26-05-2015 09-09-2015
2016	18.7±0.5 21.9±0.8	16.4±0.5 18.1±0.7	11.5±0.5 16.7±0.6	15.4±0.5 16.3±0.7	21.0±0.5 18.2±0.7	12-05-2016 06-09-2016
2017	23.3±1.9 22.4±1.9	22.7±0.8 21.9±1.0	21.3±1.0 11.1±0.7	18.8±1.5 20.0±1.0	28.3±1.2 22.6±1.0	30-05-2017 27-09-2017
2018	24.4±1.0 21.4±1.0	19.6±0.9 19.7±0.9	11.9±0.6 13.9±0.7	21.0±0.9 17.1±0.8	18.4±1.5 12.1±0.6	26-06-2018 18-10-2018
2019	22.6±1.8 27.3±1.2	22.7±1.8 22.5±1.0	13.6±0.7 20.1±0.9	19.0±1.6 20.9±0.9	15.4±0.8 14.2±0.8	04-06-2019 17-09-2019
2020	43.0±3.5 23.1±1.9	40.8±3.3 21.9±1.8	20.1±1.7 17.0±1.4	15.8±1.3 18.5±1.5	20.8±1.7 14.6±1.2	05-05-2020 09-09-2020
2021	21.4±1.1 20.8±1.0	18.6±1.0 19.9±1.0	10.8±0.6 10.8±0.6	18.2±1.0 17.8±0.9	14.0±0.8 14.8±0.8	13-08-2021 11-09-2021
2022	22.1±0.9 22.8±1.0	18.4±0.8 18.1±0.8	9.0±0.4 9.1±0.5	18.0±0.7 15.3±0.7	13.0±0.6 13.5±0.6	24-06-2022 14-09-2022
2023	36.1±1.8 20.5±1.0	32.4±1.6 18.8±0.9	30.3±1.6 17.2±0.8	18.6±0.8 16.0±0.8	24.1±1.3 13.3±0.7	04-08-2023 13-09-2023
Mean concentration	24.3±1.3	22.4±1.1	15.9±0.8	18.0±0.9	19.0±0.9	–

Table 2. Activity concentration of ^{228}Ac in soil samples measured in 2014–2023 expressed in Bq/kg.

Actinium ^{228}Ac	D-1	D-2	D-3	D-4	D-5	Collection date
2014	23.5±0.6 23.7±0.6	18.0±0.5 20.2±0.5	10.5±0.3 14.1±0.4	16.5±0.4 16.1±0.5	15.4±0.4 16.6±0.5	21-05-2014 09-09-2014
2015	25.5±0.6 22.1±0.8	20.7±0.6 20.3±0.7	18.7±0.6 14.3±0.5	19.2±0.5 18±0.6	20.7±0.6 18.5±0.6	26-05-2015 09-09-2015
2016	23.2±0.5 24.5±0.6	20.1±0.4 21.3±0.5	13.4±0.4 18.6±0.5	18.1±0.4 19.2±0.6	21.9±0.5 18.4±0.5	12-05-2016 06-09-2016
2017	24.3±0.6 23.9±0.7	21.7±0.6 21.0±0.6	16.8±0.5 10.7±0.3	17.4±0.5 19.4±0.5	21.8±0.6 20.3±0.6	30-05-2017 27-09-2017
2018	23.9±0.6 24.1±0.6	20.5±0.6 19.0±0.5	11.1±0.3 13.1±0.4	18.6±0.5 15.9±0.4	18.3±0.5 11.9±0.4	26-06-2018 18-10-2018
2019	23.6±0.6 25.3±0.7	21.0±0.6 21.3±0.6	12.9±0.4 19.5±0.5	17.3±0.5 17.9±0.5	14.0±0.4 13.9±0.4	04-06-2019 17-09-2019
2020	23.6±0.9 23.9±0.6	20.1±0.7 20.9±0.6	11.8±0.5 16.0±0.4	13.3±0.4 16.4±0.5	18.6±0.5 13.7±0.4	05-05-2020 09-09-2020
2021	24.4±0.6 25.1±0.6	21.1±0.8 21.5±0.5	11.7±0.3 11.2±0.3	18.4±0.6 17.1±0.4	13.7±0.4 13.4±0.4	13-08-2021 11-09-2021
2022	23.8±0.6 24.7±0.8	20.3±0.5 18.6±0.6	8.4±0.2 9.4±0.3	17.3±0.4 13.6±0.4	12.3±0.3 11.9±0.4	24-06-2022 14-09-2022
2023	23.1±0.8 24.0±0.6	20.0±0.7 19.5±0.5	17.9±0.6 17.0±0.4	18.1±0.5 16.2±0.4	12.5±0.4 13.1±0.3	04-08-2023 13-09-2023
Mean concentration	24.0±0.7	20.4±0.6	13.9±0.4	17.2±0.5	16.0±0.5	-

Table 3. Activity concentration of ^{40}K in soil samples measured in 2014-2023 expressed in Bq/kg.

Potasssium ^{40}K	D-1	D-2	D-3	D-4	D-5	Collection date
2014	588±13 618±14	506±13 562±13	383±9 426±10	453±12 475±11	388±9 424±10	21-05-2014 09-09-2014
2015	598±13 533±12	535±7 495±11	488±6 402±9	491±11 434±7	394±5 387±5	26-05-2015 09-09-2015
2016	547±7 569±13	504±7 534±12	381±9 421±11	462±6 483±13	400±5 406±9	12-05-2016 06-09-2016
2017	596±34 569±34	538±20 528±20	411±16 343±13	461±26 477±18	401±15 442±17	30-05-2017 27-09-2017
2018	578±21 567±21	491±19 460±17	336±13 387±15	468±18 404±15	444±26 358±14	26-06-2018 18-10-2018
2019	573±33 596±23	538±31 533±20	371±14 411±16	456±26 453±17	390±15 398±15	04-06-2019 17-09-2019
2020	587±33 572±33	543±31 546±31	363±21 410±23	398±23 430±25	478±27 388±22	05-05-2020 09-09-2020
2021	517±13 572±13	526±15 535±12	362±8 329±7	469±13 430±10	392±11 386±9	13-08-2021 11-09-2021
2022	541±11 566±16	497±10 481±12	280±6 301±7	417±9 360±9	338±7 345±8	24-06-2022 14-09-2022
2023	551±12 557±12	497±11 509±11	473±10 433±9	458±9 421±9	370±8 380±8	04-08-2023 13-09-2023
Mean concentration	570±19	518±16	386±12	445±14	395±12	-

Fig. 2–5 show the graphs of ^{226}Ra, ^{228}Ac, ^{40}K and ^{137}Cs activity concentrations depending on the sampling point. Data for the graphs were taken from three randomly selected years.

In Fig. 2–4, the activity concentrations of natural isotopes do not show significant variations between the individual sampling points (particularly ^{228}Ac and ^{40}K). Slight fluctuations can be observed in the activity concentrations of ^{226}Ra; however, when considering a 10-year dataset, mean activity concentrations can be determined by averaging the results from the five sampling points. For ^{137}Cs, the situation is somewhat different. Its activity concentrations may vary significantly depending on the sampling location; therefore, in this case, successive data series correspond to different measurement points.

Since caesium is present in the environment as a result of human activities, this study presents the deposition of caesium in the 10 cm soil layer for the analysed sampling points D-1 to D-5.

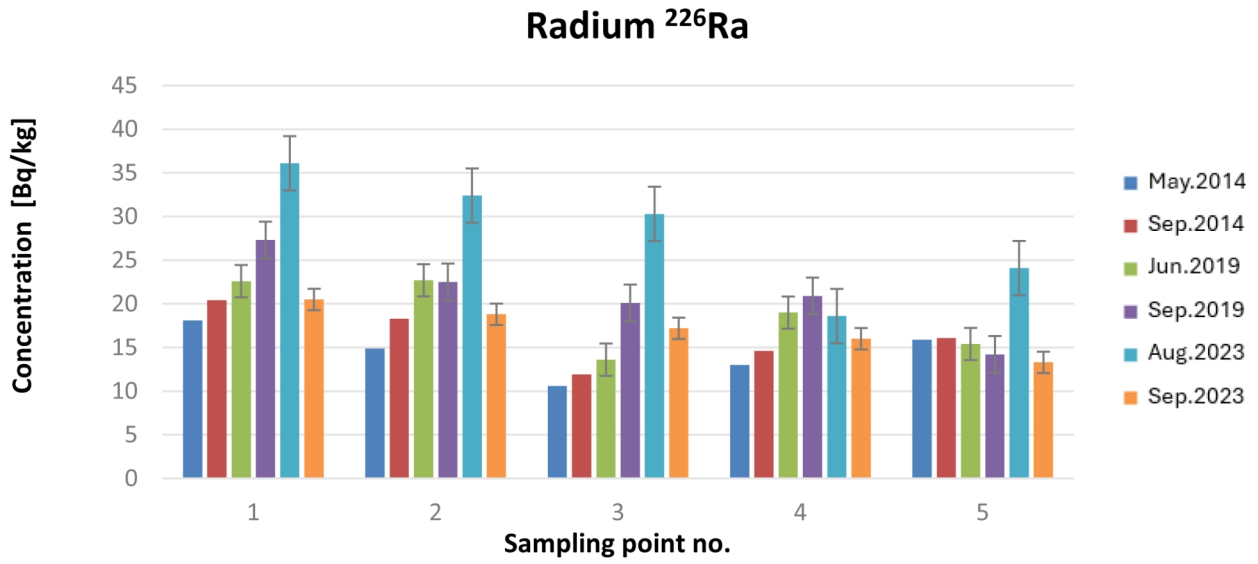

Fig. 2. Activity concentration of radium ^{226}Ra in individual soil sampling points in three randomly selected years: 2014, 2019 and 2023.

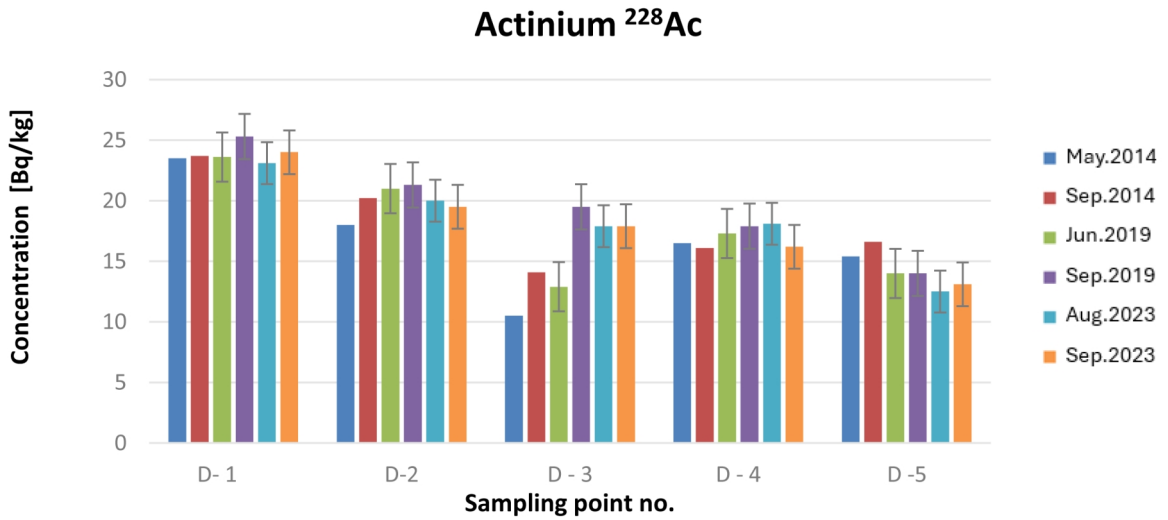

Fig. 3. Activity concentration of actinium ^{228}Ac in individual soil sampling points in three randomly selected years: 2014, 2019 and 2023.

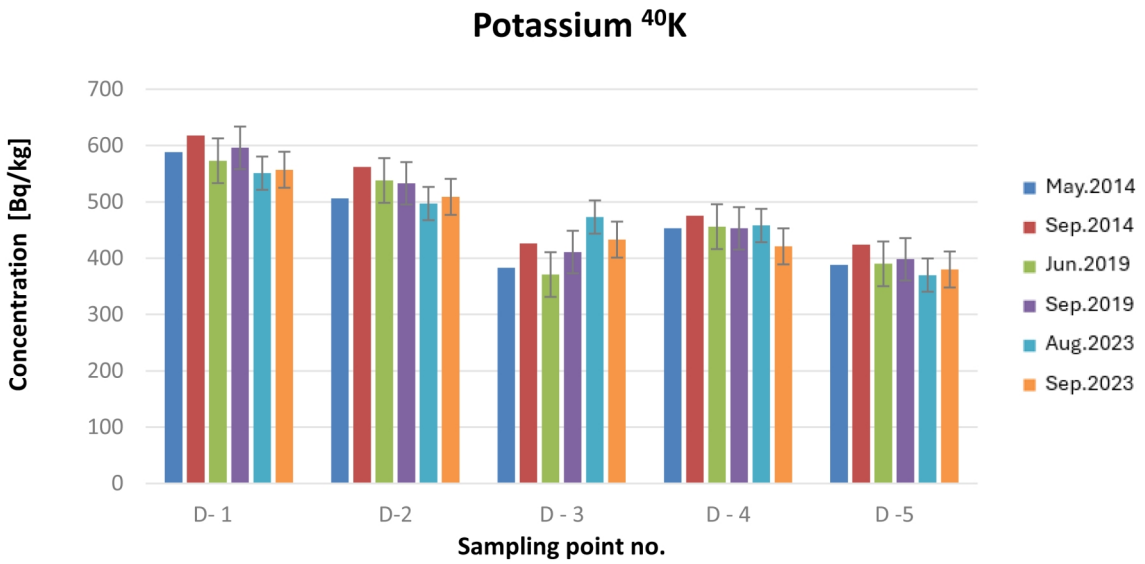

Fig. 4. Activity concentration of potassium ^{40}K in individual soil sampling points in three randomly selected years: 2014, 2019 and 2023.

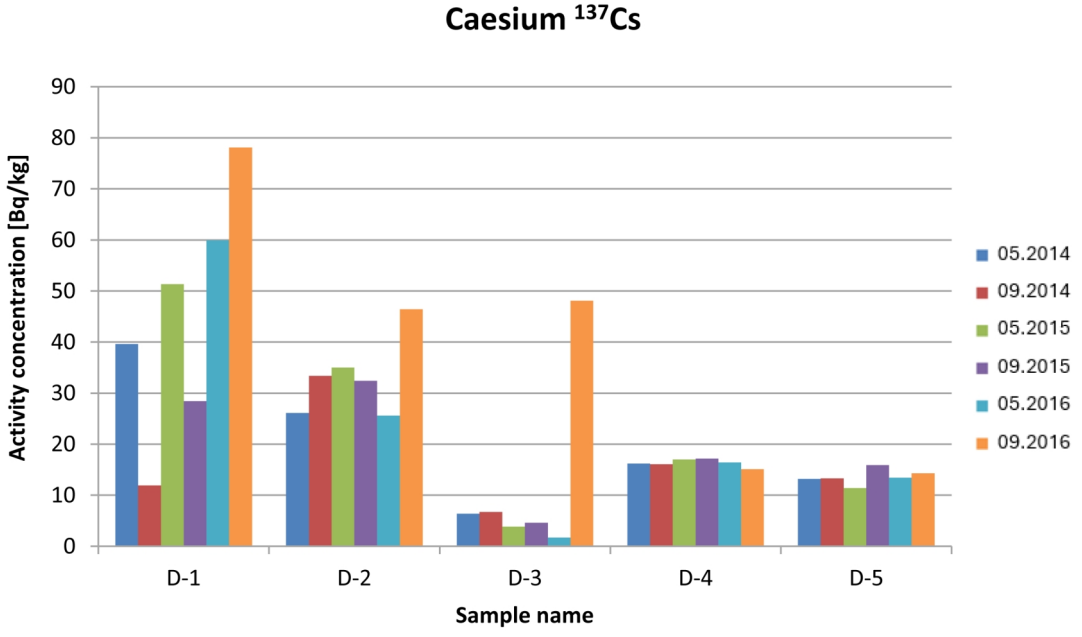

Fig. 5. Activity concentration of caesium ^{137}Cs in individual soil sampling points in the years 2014–2016.

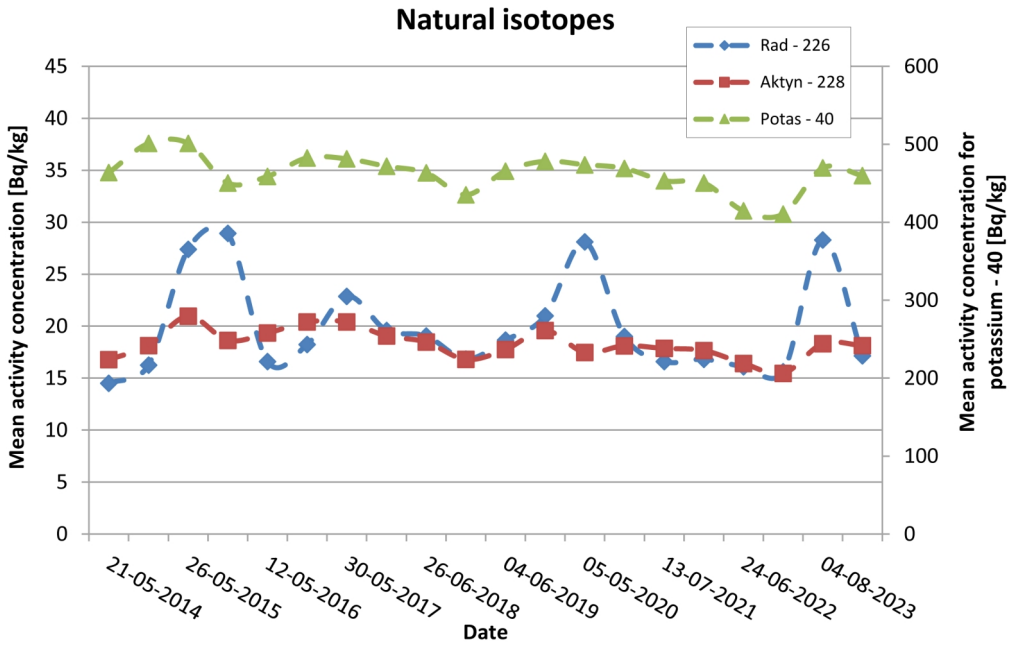

Fig. 6. Mean activity concentration of potassium ^{40}K, radium ^{226}Ra and actinium ^{228}AC in soil samples in the years 2014–2023.

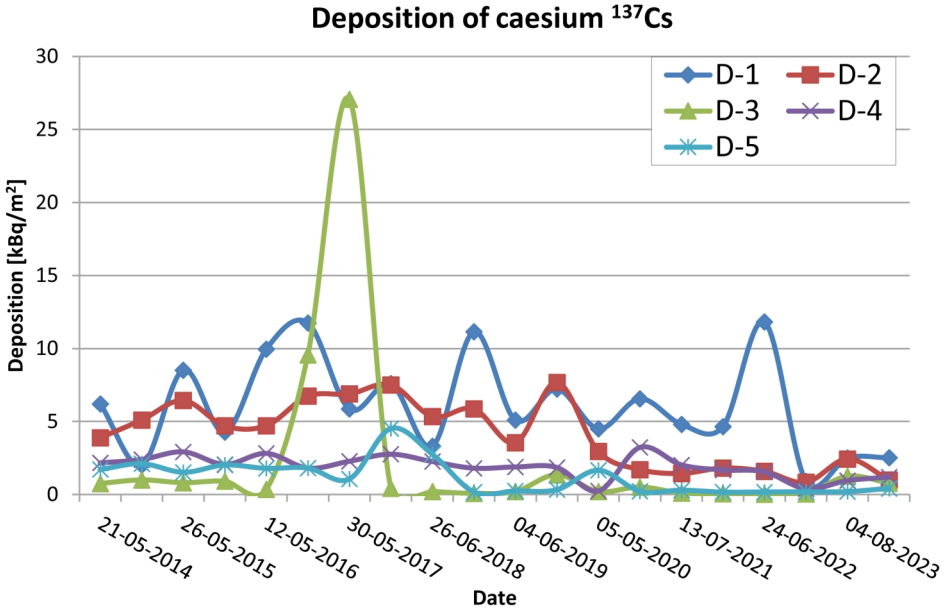

Fig. 7. Deposition of caesium ^{137}Cs in the years 2014-2023 in subsequent sampling points.

The activity concentration of ^{137}Cs was converted to deposition, expressed in kBq/m^2, using the following formula:

$$Dep\ \frac{kBq}{m^2} \quad \frac{S\ \frac{Bq}{kg}}{1000} \quad \frac{M[kg]}{7\ \ P[m^2]} \quad \frac{S\ \ M}{1750\ \ \ \ D^2} \quad \frac{kBq}{m^2}$$

where: $S\ \frac{Bq}{kg}$ is the activity concentration of caesium ^{137}Cs,

$M[kg]$ is the total mass of the sample collected, and

$P\ \ \frac{D^2}{4}$ is surface area from which soil is collected using a sampler of diameter D.

In 2022, two additional artificial radioactive isotopes were detected, namely americium ^{241}Am and cerium ^{141}Ce.

Americium was detected only in the sample collected from point D-1. Its activity concentration was 0.88±0.22 Bq/kg in the sample taken on 24 June 2022, whereas in the sample collected on 14 September 2022 its activity concentration was below the detection limit of 0.58 Bq/kg.

The artificial isotope cerium ^{141}Ce was detected in samples from points D-2, D-3 and D-4. Its activity concentrations are presented in Table 5.

Table 4. Deposition of caesium ^{137}Cs in soil samples from subsequent sampling points, expressed in kBq/m^2.

Caesium ^{137}Cs	D-1	D-2	D-3	D-4	D-5	Collection date
2014	6.20±0.11 2.02±0.03	3.89±0.10 5.10±0.09	0.76±0.01 1.00±0.02	2.17±0.05 2.40±0.04	1.71±0.03 2.11±0.05	21-05-2014 09-09-2014
2015	8.53±0.15 4.27±0.08	6.45±0.04 4.70±0.09	0.81±0.01 0.91±0.02	2.92±0.05 2.06±0.01	1.52±0.01 2.02±0.01	26-05-2015 09-09-2015
2016	9.95±0.05 11.76±0.20	4.71±0.04 6.74±0.12	0.36±0.01 9.56±0.20	2.82±0.02 1.81±0.05	1.79±0.01 1.82±0.04	12-05-2016 06-09-2016
2017	5.86±0.13 7.60±0.20	6.89±0.14 7.52±0.15	27.07±0.47 0.43±0.02	2.28±0.06 2.76±0.07	1.06±0.02 4.52±0.08	30-05-2017 27-09-2017
2018	3.31±0.07 11.15±0.22	5.34±0.11 5.87±0.11	0.21±0.01 0.09±0.01	2.26±0.05 1.80±0.03	2.76±0.07 0.18±0.07	26-06-2018 18-10-2018
2019	5.10±0.12 7.23±0.14	3.55±0.08 7.69±0.15	0.21±0.01 1.36±0.03	1.89±0.05 1.85±0.04	0.25±0.01 0.33±0.01	04-06-2019 17-09-2019
2020	4.50±0.10 6.56±0.15	2.95±0.07 1.71±0.04	0.21±0.01 0.52±0.01	0.25±0.01 3.23±0.07	1.66±0.04 0.22±0.01	05-05-2020 09-09-2020
2021	4.81±0.10 4.66±0.08	1.44±0.04 1.82±0.04	0.11±0.01 0.05±0.01	2.03±0.05 1.68±0.04	0.30±0.01 0.17±0.01	13-08-2021 11-09-2021
2022	11.82±0.17 0.90±0.02	1.59±0.03 0.84±0.02	0.03±0.01 0.04±0.01	1.56±0.02 0.40±0.02	0.18±0.01 0.19±0.01	24-06-2022 14-09-2022
2023	2.50±0.05 2.52±0.05	2.44±0.04 0.99±0.02	1.25±0.02 0.79±0.02	0.95±0.02 1.19±0.02	0.20±0.01 0.42±0.01	04-08-2023 13-09-2023

Table 5. Activity concentration of cerium ^{141}Ce in soil samples measured in 2022 expressed in Bq/kg.

Cerium ^{141}Ce	D-2	D-3	D-4
24-06-2022	0.32±0.06	0.15±0.04	0.38±0.06
14-09-2022	0.79±0.13	0.20	0.25

Results analysis

The activity concentration of potassium ^{40}K in 2014–2023 ranged from 280 Bq/kg recorded in 2022 for point D-3 to 618 Bq/kg obtained in 2014 in point D-1. The activity concentration of radium ^{226}Ra ranged from 9 Bq/kg recorded in 2022 at point D-3 to 43 Bq/kg in 2020 at point D-1, while in the case of actinium ^{228}Ac the activity concentration ranged from approx. 8.4 Bq/kg in 2022 in point D-3 to 25.5 Bq/kg in 2015 in point D-1.

The level of activity concentration of natural isotopes remains fairly constant, however, some peaks can be seen on the graph of radium ^{226}Ra. However, these variations fall within the typical range for this radium isotope, and therefore no particular attention was devoted to them.

The levels of activity concentrations of natural isotopes do not differ significantly from the mean values for Poland. The mean concentration of radium ^{226}Ra in Poland is: 31±1.6 Bq/kg in the range from 6.4 to 154.7 Bq/kg, while in the Mazowieckie Voivodeship, where the KSOP is located, it is 15.9 Bq/kg. In the case of actinium ^{228}Ac, in Poland the mean concentration is 30.5 Bq/kg in the range from 6 to 129.2 Bq/kg, and in the Mazowieckie Voivodeship the mean concentration is 15.9 Bq/kg. The mean activity concentrations for the Mazowieckie Voivodeship are exceeded in certain places, but due to the fact that the concentration levels oscillate around certain mean values, the same point where the measurements were carried out exceeds this mean level once, and in the following year it is below the mean value. Such variations are typical for soil, as radioactive isotopes migrate with soil material and their levels fluctuate over time.

In the case of potassium ^{40}K, the mean activity concentration in Poland is 497±15 Bq/kg in the range from 138 to 1046 Bq/kg, where in the Mazowieckie Voivodeship the mean concentration is 382 Bq/kg. It is the only isotope that exceeds the mean concentration in the Mazowieckie Voivodeship for each measurement point; however, it exceeds it by values that can be attributed to a different soil structure.

The mean activity concentration of natural isotopes in the Mazowieckie Voivodeship is lower than the mean concentration for the whole of Poland, which can also be

seen in the measured mean concentrations of radium and actinium, which were lower than the mean concentrations in Poland. The exception here is potassium, the concentration of which is close to national concentrations and exceeded the mean concentrations in the Mazowieckie Voivodeship.

In the case of caesium ^{137}Cs deposition, significant fluctuations in time and a large dependence of the deposition value on the examined point are visible. This suggests that in some points, i.e. those where this level is quite constant, the caesium content in the soil is the result of older events, e.g. the accident at the Chernobyl nuclear power plant, while the points where the deposition is greater (point D-1) and variable over time (point D-3) may suggest that „excess" caesium comes from a repository.

The mean value of caesium ^{137}Cs deposition in Poland is 1.96±0.19 kBq/m^2 in the range from 0.20 to 16.63 kBq/m^2, while in the Mazowieckie Voivodeship the mean deposition is 1.58±0.39 kBq/m^2 in the range from 0.25 to 7.09 kBq/m^2, so the mean value is lower for the Mazowieckie Voivodeship than that from the whole country. Considering that this isotope of caesium does not occur naturally, we can expect higher values of deposition in concentrated areas, which may be caused, for example, by local rainfall from clouds contaminated after the Chernobyl nuclear power plant accident or contamination resulting, for example, from a damaged caesium source. The values of deposition in the KSOP environment are generally greater than the mean values for both Poland and the Mazowieckie Voivodeship, however, these are still values that do not have a significant negative impact on people and the environment.

Summary

Radiological monitoring of the soil in the proximity of the National Radioactive Waste Repository in Różan showed the presence of natural isotopes in all five samples, i.e. potassium ^{40}K, radium ^{226}Ra and actinium ^{228}Ac and one isotope of artificial origin – caesium ^{137}Cs. The activity concentrations of natural isotopes remain constant and are related to the geological structure of the area. However, the presence of caesium ^{137}Cs deposition caused by human activity remains low. The calculated deposition of ^{137}Cs in the analysed soil samples is comparable to the deposition of this isotope obtained for the soil of the Mazowieckie Voivodeship (with the exception of point D-1), which proves the possibility of its origin at the time of the Chernobyl nuclear power plant accident [2].

Taking into account the above tests, the presence of the Radioactive Waste Repository in Różan does not adversely affect the surroundings of the said location in terms of radioactive gamma.

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