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^{137}Cs ACTIVITY CONCENTRATION IN THE GROUND-LEVEL AIR IN THE IGNALINA NPP REGION

R. Jasiulionis and A. Rožkov

Institute of Physics, Savanorių 231, LT-02300 Vilnius, Lithuania
E-mail: rimjas@ktl.mii.lt, rozkov@ar.fi.lt

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Results of the ^{137}Cs activity concentration measurements in the ground-level air in the Ignalina Nuclear Power Plant (Ignalina NPP) region in 1978–2006 are presented and discussed. The peculiarities of the trend of mean annual radiocaesium activity concentrations in the air during the studied period are explained and described empirically. The analysis of the radiocaesium concentration course after the Chernobyl accident shows that mean annual ^{137}Cs activity concentrations in the air are decreasing slower than it is expected due to its radioactive decay and aerosol, as the ^{137}Cs carrier, removal processes in the atmosphere. The Hybrid Single-Particle Lagrangian Integrated Trajectory model of the global dispersion and deposition of the admixture in the air is used to study episodic increases in the ^{137}Cs activity concentration in the ground-level air of more than 8 μBq m^{-3}. Modelling results show the transport of resuspended radiocaesium with air mass from the Chernobyl NPP accident polluted regions to be an important source of the radionuclide to the environment of the Ignalina NPP. It is shown that ^{137}Cs emissions from its generation sources (nuclear industry objects) and, probably, the depositional flux of the radiocaesium from the stratospheric reservoir, can contribute up to 10% to the mean annual ^{137}Cs activity concentration value. Mean arithmetic and mean geometric ^{137}Cs activity concentrations in the air during a year are calculated and compared.

Keywords: ^{137}Cs, activity concentration, ground-level air, Ignalina NPP, HYSPLIT model

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1. Introduction

The environment around the Ignalina Nuclear Power Plant (Ignalina NPP) has been a part of the radioecological monitoring carried out at the Ignalina Radioecological Monitoring Station of the Institute of Physics (PhI station) since 1978 [1, 2]. Analysis of permanent measurements of the radionuclide activity concentrations in the ground-level air and deposition in the Ignalina NPP region, using radionuclides as tracers of air masses transport, gives insight into understanding of the radionuclide behaviour in the Ignalina NPP environment.

^{137}Cs ($t_{1/2} = 30.07$ years) is formed by the radioactive decay of its progenitor nuclides ^{137}I , ^{137}Xe, the uranium fission products, as follows:

$$^{137}\mathrm{I} \overset{\beta^-}{\longrightarrow} {}^{137}\mathrm{Xe} \overset{\beta^-,\gamma}{\longrightarrow} {}^{137}\mathrm{Cs}\,.$$

^{137}Cs is one of the most distributed artificial radionuclides in the environment, and its activity concentrations in the air are still registered at the radioecological monitoring stations in the world [3].

Large quantities of ^{137}Cs were released into the environment in the 20th century during nuclear weapon tests and accidents at the nuclear industry objects, the most important of which were the accidents at the Windscale NPP (the UK, 1957), the Kyshtym radioactive waste storage (the USSR, 1957), the nuclear fuel reprocessing plant "Majak" (the USSR, 1967), the Three Mile Island NPP (the USA, 1979) and the Chernobyl NPP (the USSR, 1986) [4].

The amount of fission products released into the air, as a result of about 500 nuclear and thermonuclear tests in the atmosphere in 1945–1980, is estimated to be $1.8{\cdot}10^{21}$ Bq. During the nuclear explosion, radionuclides are released into the troposphere, they become attached to the atmospheric aerosol, and can be transported with air masses over long distances, usually along the latitude at which the original test was performed, and are removed by such processes as wash-out with the precipitation, dry deposition, vertical transport to the ground with turbulent and convectional air mass fluxes. During thermonuclear explosions, radionuclides condense in the stratosphere, become distributed all over the world with the moving stratospheric

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air masses, and later get into the troposphere causing the radionuclide global fallout. The ^{137}Cs total emission into the stratosphere during nuclear and thermonuclear weapon tests is estimated to be $9.5{\cdot}10^{15}$ Bq [5].

During the accident at the Chernobyl NPP Unit 4, about $8.5{\cdot}10^{16}$ Bq of radiocaesium was released into the troposphere. More than half of ^{137}Cs was deposited in the vicinity of the destroyed reactor, and another part was transported with air masses over large distances causing its fallout in the world. The total area of contamination with the Chernobyl-origin ^{137}Cs with the density of 1–5 Ci km^{-2} is estimated to be about 150 thousand km^2 [6].

There were 435 nuclear power units of the total installed capacity of 370 GW in operation in the world at the beginning of 2007 [7]. The annual total particulate release from the nuclear industry objects is estimated to be about $8.5{\cdot}10^{10}$ Bq GW^{-1} [8]. Thus, nowadays the emissions from operating nuclear industry objects can be treated as an additional source of radiocaesium to the environment that cannot be neglected.

The study of the ^{137}Cs dynamics in the ground-level air and its deposition over a long period of time can help much in understanding the radionuclide removal processes in the atmosphere and provide information about its sources in the past and at present.

The aim of this work was to explain the peculiarities of the trend of mean annual ^{137}Cs activity concentrations in the ground-level air in 1978–2006 and evaluate the impact of present sources of ^{137}Cs on its activity concentrations in the air in the Ignalina NPP region.

2. Method

In this study the term "Ignalina NPP region" means a 30 km zone around the assumed centre of the Ignalina NPP Units 1 and 2 (N 55° 36′ 17.7″, E 26° 33′ 44.1″).

The air filtration equipment for continuous collection of air-borne radionuclides is installed at the PhI station (N 55° 34′ 34.0″, E 26° 35′ 25.7″) situated at a 3.5 km distance southeast from the Ignalina NPP. The high airflow rate of about 1800 m^3 h^{-1} through the perchlorvinyl Petrianov filters FPP-15-1.5 with an area of 0.9 m^2 was used for aerosol sampling once every three days till 1998 and later once a week. Filters were pressed into pellets of the standard geometry (diameter 40 mm, height 15–25 mm), and activities of radionuclides were measured by the gamma spectrometry method. The minimum ^{137}Cs detectable activity of 0.1 μBq m^{-3} was calculated for the 80 000 s live measurement time (the pellet of 20 mm height, the sampled air volume 200 000 m^3).

Atmospheric precipitation samples were collected at the PhI station: rain samples from the 10 m^2 sloping surface into the 80 l bath, and snow samples from the 1–2 m^2 surface near the PhI station. A method of rapid precipitation with the iron, cobalt, and manganese hydroxides was used to concentrate radionuclides in deposition samples [9]. Particles in the melted snow or rain sample were separated using the 0.1 mm diameter pore filter. The filtrate was treated radiochemically by slightly acidifying the solution, adding iron (III), cobalt (II), manganese (II) chlorides and then ammonia, and finally adjusting pH $\cong$ 9. Then the sample was well mixed, left to stay overnight, the solution was decanted, and deposits of metal hydroxides were separated. The decanted solution was additionally evaporated and the yield of the radionuclide in separated deposits was calculated from the ratio between the radionuclide activity detected in deposits and that of the total radionuclide activity in deposits and the decanted solution. The ^{137}Cs yield was about 20%. The minimum ^{137}Cs detectable activity in deposits of 50 mBq was calculated for the 80 000 s live measurement time (the pellet of 20 mm height).

The ORTEC portable gamma spectrometer with the coaxial high-purity germanium (HPGe) detector (relative efficiency 30%, 1.85 keV/1.33 MeV) and a passive shield of lead bricks with cadmium and copper layers was used for the gamma spectrometry. The SNIP (SILENA) multichannel analyzer was used to obtain energy spectra of radionuclides. The radionuclide net count rate in a selected photopeak (in cps) and the statistical measurement error (1σ standard deviation) were calculated using the EMCAPLUS software. The ^{137}Cs activity concentration was calculated from its daughter ^{137m}Ba photopeak (661.7 keV) in respect of the gamma emission probability, gamma registration efficiency in a given geometry, and sample volume.

The mean arithmetic radiocaesium activity concentration in the air during a year was calculated including sampling periods with the ^{137}Cs activity concentration in the air below its detection limit.

The trend of the mean annual ^{137}Cs activity concentration in the ground-level air for the selected period of time was approximated using the ORIGIN software as follows:

$$A = A_0\, \mathrm{e}^{\Lambda(t-t_0)}\,, \tag{1}$$

where A is the radionuclide activity concentration in the air (μBq m^{-3}) in a particular year t, A_0 is the radionu-

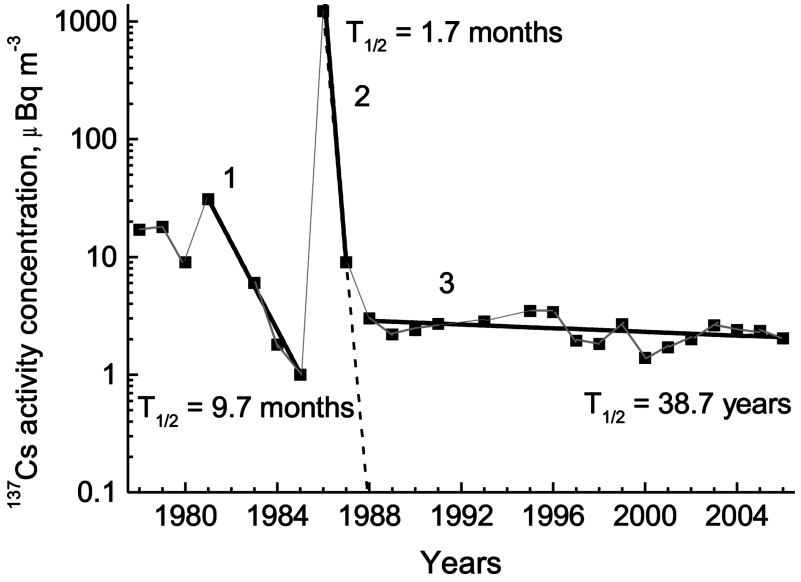

Fig. 1. Mean annual ^{137}Cs activity concentrations in the ground-level air in the Ignalina NPP region in 1978–2006.

clide activity concentration (μBq m^{-3}) at the initial moment of time t_0 (year), and Λ is the empirical constant of the radionuclide removal from the ground-level air (y^{-1}).

The half-time of the ^{137}Cs removal from the ground-level air $T_{1/2}$ (y) was calculated as

$$T_{1/2} = \frac{\ln 2}{\Lambda}\,. \qquad (2)$$

The 72-hour backward trajectories of air mass transport to the PhI station for certain aerosol sampling periods were calculated using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model [10]. The Greenwich Mean Time was used in calculations.

Data on the ^{137}Cs emission rate from the Ignalina NPP and the amount of atmospheric precipitation in the region were provided by the Ignalina NPP Radiation Protection Laboratory.

3. Results and discussion

3.1. The trend of mean annual ^{137}Cs activity concentrations in the ground-level air in 1978–2006

The data of permanent measurements of ^{137}Cs activity concentrations in the ground-level air in the Ignalina NPP region in 1978–2006 were analysed. Three periods with a different mean annual radiocaesium activity concentration trend can be distinguished indicating different ^{137}Cs sources in the air (Fig. 1).

The radiocaesium dynamics in the ground-level air in 1978–1985 was determined by its global fallout from the stratospheric reservoir caused by nuclear and thermonuclear weapon tests in the atmosphere. The increase in the mean annual ^{137}Cs activity concentration in the air up to 31 μBq m^{-3} in 1981 was a result of the last Chinese nuclear test in the atmosphere performed on October 16, 1980 as well as of the previously accumulated ^{137}Cs in the stratosphere. The half-time of the radionuclide removal in the ground-level air during 1981–1985 ($T_{1/2}$ = 9.7 months) (Fig. 1, graph *1*) was calculated, which evidently shows the global fallout of the radionuclide from the stratospheric reservoir to be the main source of radiocaesium to the ground-level air till 1986.

The ^{137}Cs emission to the atmosphere during the accident at the Chermobyl NPP in 1986 resulted in the increase of its mean annual activity concentration in the air up to 1230 μBq m^{-3}. The relatively short half-time of the radionuclide removal in the ground-level air during 1986–1988 ($T_{1/2}$ = 1.7 months) indicates the radionuclide fallout from the troposphere (Fig. 1, graph *2*).

Obviously, after the Chernobyl accident the radiocaesium activity concentrations in the ground-level air, assuming no significant radionuclide emission to the atmosphere since 1986, should be decreasing due to its radioactive decay and aerosol, as the ^{137}Cs carrier, removal processes in the atmosphere. However, since 1988 the observed mean annual ^{137}Cs activity concentration values have been decreasing slower than it was expected – the half-time of the radionuclide removal in the ground-level air was calculated to be 38.7 years (Fig. 1, graph *3*). It can be explained either by the sta-

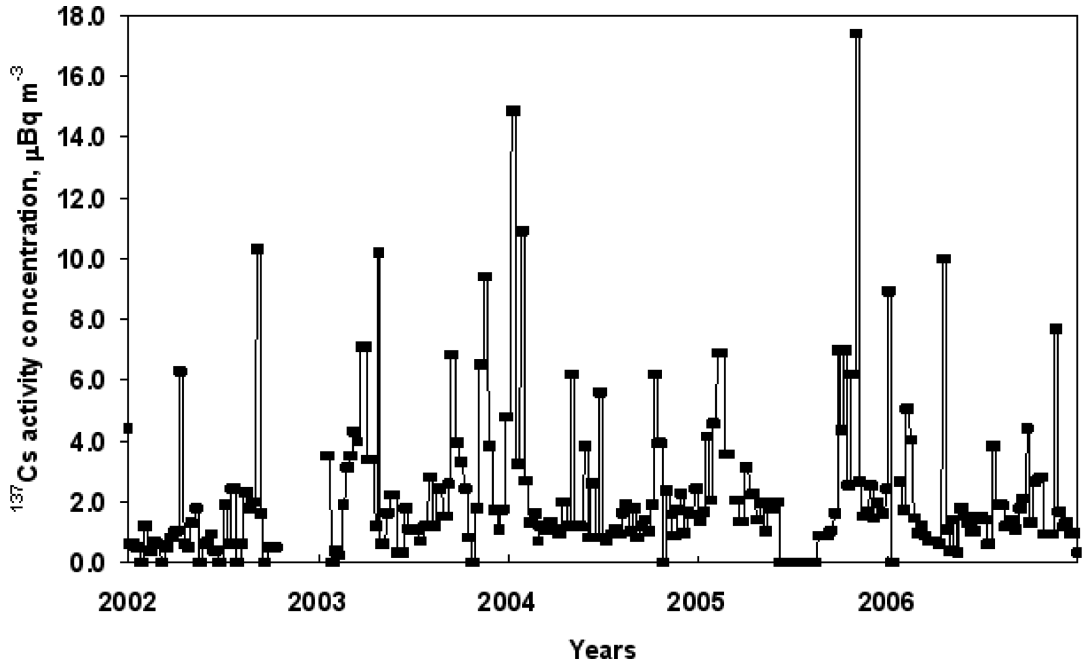

Fig. 2. The ^{137}Cs activity concentrations in the ground-level air in the Ignalina NPP region in 2002–2006.

tionary process of the aerosol, as the radionuclide carrier, resuspension and deposition in the environment or by the existence of an additional radiocaesium source to the ground-level air, e. g., emissions from world nuclear industry objects.

These results of the mean annual ^{137}Cs activity concentration analysis agree with those performed at air monitoring stations in Germany [11], Greece [12], and Russia [13].

The evaluation of present radiocaesium sources to the atmosphere after the Chernobyl accident would help in understanding the dynamics of ^{137}Cs in the environment nowadays.

3.2. Sources of ^{137}Cs in the air after the Chernobyl accident

Episodic increases in weekly ^{137}Cs activity concentrations during certain aerosol sampling periods were registered at the PhI station after the Chernobyl accident. The results of weekly measurements of the ^{137}Cs activity concentration in the ground-level air in the Ignalina NPP region in 2002–2006 are presented in Fig. 2.

It is shown in Fig. 2 that annual mean arithmetic ^{137}Cs activity concentrations can be influenced by the radiocaesium concentration increases over certain periods. Annual mean activity concentrations of the globally distributed ^{137}Cs at the latitude of the PhI station are better estimated using the radiocaesium mean geometric activity concentration values.

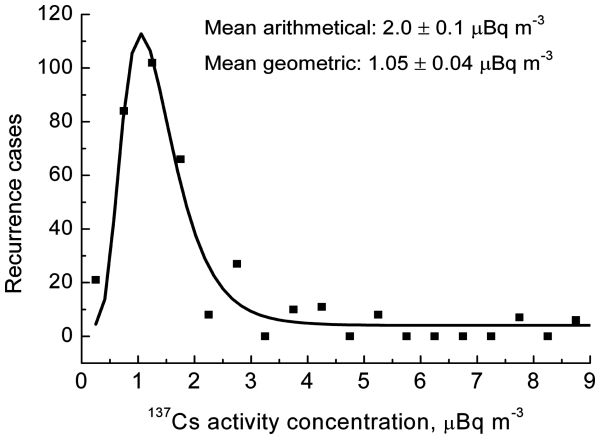

Fig. 3. The lognormal distribution of weekly ^{137}Cs activity concentrations in the ground-level air in the Ignalina NPP region in 2006.

The distribution of the radiocaesium activity concentrations in the air in 1978–1980 is satisfactorily described by the truncated normal distribution and that in 2004–2006 by the lognormal distribution. An example of the lognormal distribution of ^{137}Cs activity concentrations in 2006 is given in Fig. 3.

The annual mean geometric ^{137}Cs activity concentration in the air in the period of 1978–1980 was 2.0 μBq m^{-3}. The annual mean geometric ^{137}Cs activity concentration in the air in the period of 2004–2006 was 1.2 μBq m^{-3}, and its mean arithmetic concentration was 2.4 μBq m^{-3}.

The seasonal maxima of the radionuclide activity concentrations were detected in April and September, and the minima in June and July, which can be ex-

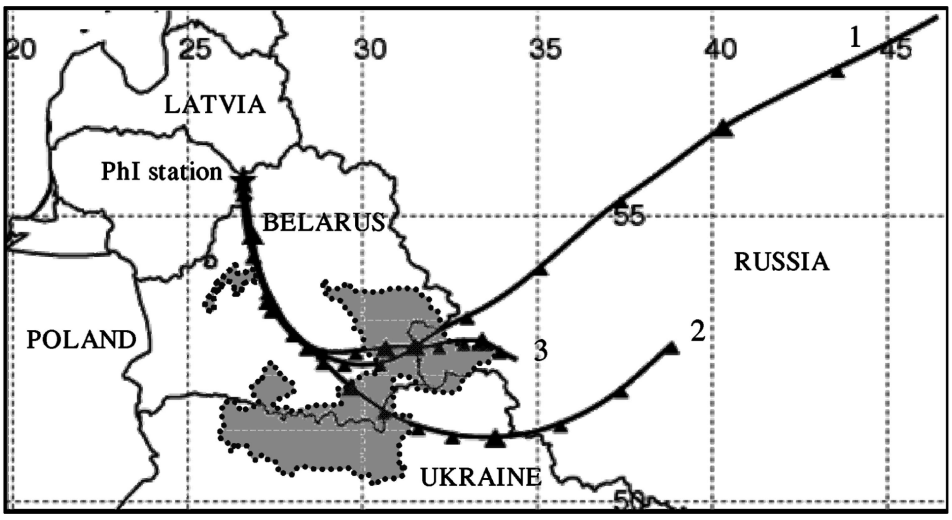

Fig. 4. Backward trajectories of air mass transport to the Ignalina NPP region that coincide with the aerosol sampling period of October 30–November 5, 2005 at the PhI station. *1* is the air mass backward trajectory ending on October 30, 2005 12:00 at the PhI station, *2* is ending on November 1, 2005 6:00, and *3* is ending on November 5, 2005 12:00. The contour shows territories of the ^{137}Cs surface contamination of more than 40 kBq m^{-2}.

plained by the enhanced resuspension of the aerosol during agricultural activities and a different amount of atmospheric precipitation during these months. In April and September the total amount of precipitation was 20 and 31 mm, respectively, and in June–July it was 73 mm, which sufficiently washes out the ground-level air aerosol, as the carrier of ^{137}Cs.

Using the HYSPLIT model, backward trajectories of air mass transport to the Ignalina NPP region were calculated for the cases of episodic increases of the ^{137}Cs activity concentration in the air in 2002–2006.

Backward trajectories of the air mass transport to the PhI station for the air sampling period of October 30–November 5, 2005, during which the ^{137}Cs activity concentration of 17.4 μBq m^{-3} has been registered, are presented in Fig. 4. The air mass backward trajectory ending on November 1, 2005 6:00 is given to show the amplitude of the change in the air mass movement during the calculation time.

The 72-hour backward trajectories of air mass transport to the PhI station (10 m above ground level) were calculated every 6 hours for aerosol sampling periods, during which the increase of the ^{137}Cs activity concentration in the air of more than 8 μBq m^{-3} was registered. Backward trajectories of air masses that passed over the territories contaminated with ^{137}Cs by more than 40 kBq m^{-2} after the Chernobyl accident [14] were selected. The ratio between the selected and the total number of trajectories was calculated. This ratio shows the probability $P_{\rm C}$ that the resuspended aerosol could be transported to the PhI station from the Chernobyl accident polluted regions (Table 1). The probability $P_{\rm I}$ of the possible aerosol transport in a puff, emitted from the Ignalina NPP, to the PhI station was calculated as well.

Table 1. Radionuclide activity concentrations in the ground-level air and probabilities of the air mass transport to the PhI station from the Chernobyl accident polluted regions ($P_{\rm C}$) or Ignalina NPP region ($P_{\rm I}$) during the indicated aerosol sampling period at the PhI station.

Sampling period	Activity concentration, μBq m^{-3}				$P_{\rm C}$	$P_{\rm I}$
	^{137}Cs	^{7}Be *	^{60}Co	^{54}Mn		
2002.09.03–10	10.3±1.8	2.7±0.3	<0.1	<0.1	0.46	0.07
2003.04.26–30	10.2±1.6	2.1±0.2	<0.1	<0.1	0.15	0
2003.11.15–23	9.4±1.7	2.9±0.3	<0.1	<0.1	0.38	0.09
2004.01.06–16	14.9±2.7	7.5±0.6	2.8±0.7	<0.1	0.38	0.03
2004.01.25–02.02	10.9±1.6	3.1±0.2	<0.1	1.0±0.3	0.75	0
2005.10.30–11.05	17.4±3.5	8.3±0.7	<0.1	<0.1	1.00	0
2005.12.31–2006.01.07	8.9±1.5	1.1±0.1	<0.1	<0.1	0.68	0
2006.04.15–23	10.0±1.8	13.5±1.1	<0.1	<0.1	0.06	0.22

* mBq m^{-3}

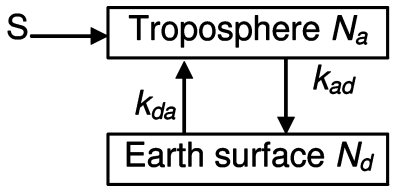

Fig. 5. The scheme of ^{137}Cs flux two-box model. N_a is the radionuclide inventory in the troposphere (Bq), N_d is the inventory of the radionuclide accumulated on the Earth surface (Bq), k_{ad} is the rate constant (y^{-1}) of the radionuclide depositional flux from the troposphere to the Earth surface, and k_{da} is the rate constant (y^{-1}) of the radionuclide resuspension flux from the Earth surface to the troposphere. S is the inflow of the radionuclide from its production sources (nuclear industry objects) and, probably, the stratospheric reservoir (Bq).

In most cases, episodic increases in the ^{137}Cs activity concentration in the air in the Ignalina NPP region correlate with the probability of the air mass movement over the ^{137}Cs contaminated territories of the Ukraine, Belarus, and Russia. On April 26–30, 2003, the air mass was moving over the Leningrad NPP. The ^{137}Cs concentration increase in the air on April 15–23, 2006 can be explained by the impact of the Ignalina NPP. However, the impact of the Ignalina NPP can be treated as insignificant in most cases.

These results show that transport of the resuspended aerosol, as the carrier of ^{137}Cs, with air mass from the Chernobyl accident polluted areas can contribute to the radiocaesium activity concentrations in the air in the Ignalina NPP region. Since this process is strongly dependent on meteorological conditions, it results in episodic radiocaesium activity concentration increases during certain periods of time.

3.3. Evaluation of additional ^{137}Cs emission sources in the ground-level air

The change in the mean annual ^{137}Cs activities in the troposphere at the latitude of the PhI station after the Chernobyl accident can be introduced using a simplified two-box model (Fig. 5).

Thus, the change in mean annual ^{137}Cs activities in the troposphere can be described as follows:

$$\frac{\mathrm{d}N_a}{\mathrm{d}t} = -(k_{ad}+\lambda)N_{a0} + (k_{da}+\lambda)N_{d0} + \lambda S\,, \quad (3)$$

where N_a is the radionuclide inventory in the troposphere (Bq) that is subject to change over time t (y), N_{a0} and N_{d0} are the radionuclide inventories in the troposphere and the Earth surface (Bq) at the initial moment of time ($t = 0$), k_{ad} is the rate constant (y^{-1}) of the radionuclide depositional flux from the troposphere to the Earth surface, k_{da} is the rate constant (y^{-1}) of the radionuclide resuspension flux from the Earth surface to the troposphere, λ is the radionuclide radioactive decay constant (y^{-1}), S is the inflow of the radionuclide from its additional production source and, probably, stratospheric reservoir (Bq).

Assuming that the resuspension and deposition of the aerosol, as the ^{137}Cs carrier, are the most important processes of its exchange between the "troposphere"–"Earth surface" reservoirs and the vertical migration (loss) of the radiocaesium to the ground surface is unsignificant, the radionuclide depositional and resuspensional fluxes can be treated as stationary ones, and the balance between them can be established ($k_{da} = k_{ad}$). Thus, the balance equation of mean annual ^{137}Cs activities in the troposphere can be described as follows:

$$-\lambda(N_0 - S) = -\Lambda(N_0 + S)\,, \quad (4)$$

where N_0 is the total inventory of the resuspended radionuclide (Bq) in the troposphere in 1988 and Λ is the rate of the radionuclide removal from the air (y^{-1}) calculated from the mean annual ^{137}Cs activity concentration trend for the period of 1988–2006. N_0 is supposed to decrease due to the radioactive decay over time.

By solving Eq. (4), the inflow of the radionuclide from its additional production source S can be evaluated. According to calculations, the additional ^{137}Cs source would contribute up to about 10% to the total radiocaesium amount circulating in the "troposphere"–"Earth surface" reservoirs.

Results of the ^{137}Cs activity concentration measurements in the ground-level air and deposition at the PhI station [15] as well as data on the ^{137}Cs emission from the Ignalina NPP ventilation stacks were used in order to verify the impact of the radiocaesium additional emission source on the environment.

According to calculation results, using the value of the ^{137}Cs depositional flux to the ground at the PhI station of 14.5 Bq m^{-2} y^{-1} in 2005, the radiocaesium emission from its additional production sources can contribute up to 10% of mean annual ^{137}Cs activity concentrations in the ground-level air.

4. Conclusions

The half-time of the ^{137}Cs removal from the ground level air in the Ignalina NPP region has been calculated to be 9.7 months in 1978–1985, 1.7 months in 1986–1988, and later – 38.7 years. The calculated half-time of the ^{137}Cs removal from the air enables us to determine sources of the radiocaesium in the air in the past.

After the Chernobyl accident the mean annual ^{137}Cs activity concentrations in the air are decreasing much slower than it is expected due to its radioactive decay and aerosol removal processes in the atmosphere.

The transport of the resuspended aerosol, as the ^{137}Cs carrier, with air masses passing over the territories contaminated with ^{137}Cs by more than 40 kBq m^{-2} after the Chernobyl accident is an important source of radiocaesium in the air in the Ignalina NPP region causing ^{137}Cs activity concentration increases in the air of more than 8 μBq m^{-3} over certain periods of time.

It is shown that the ^{137}Cs emissions from its generation sources (nuclear industry objects) and, probably, from its stratospheric reservoir, can contribute up to 10% to the mean annual ^{137}Cs activity concentration value. This result can help in evaluating the ^{137}Cs ionization radiation doses caused by aerosol, as the ^{137}Cs carrier, emissions from its additional sources.

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^{137}Cs AKTYVUMO KONCENTRACIJA IGNALINOS AE REGIONO PAŽEMIO ORE

R. Jasiulionis, A. Rožkov

Fizikos institutas, Vilnius, Lietuva

Santrauka

Pateikiami ir aptariami ^{137}Cs aktyvumo koncentracijos pažemio ore matavimų Ignalinos AE regione 1978–2006 metais rezultatai. Vidutinės metinės ^{137}Cs aktyvumo koncentracijos ore eiga aproksimuota empirine lygtimi ir suskirstyta į tris laikotarpius: I – 1981–1985 m., kai ^{137}Cs šaltiniu buvo radionuklidai, Kinijos branduolinio ginklo bandymo ore (1980 m.) metu pasiekę stratosferą (^{137}Cs metinės vidutinės aktyvumo koncentracijos pažemio ore pusėjimo trukmė $T_{1/2}$ = 9,7 mėn.), ir jų globalios iškritos iš stratosferinio rezervuaro; II – 1986–1988 m. radionuklidų pernaša troposferoje ($T_{1/2}$ = 1,7 mėn.), įvykus Černobylio AE avarijai; III – nuo 1988 m. iki šiol, kai vidutinė metinė ^{137}Cs aktyvumo koncentracija ore mažėja lėčiau ($T_{1/2}$ = 38,7 m.), nei turėtų mažėti dėl radioaktyviojo skilimo ir atmosferos savivalos procesų. Manoma, kad tiriamuoju laikotarpiu ^{137}Cs koncentracijos kitimą lėmė aerozolio, radionuklido nešiklio, antrinis pakėlimas nuo žemės paviršiaus ir branduolinės energetikos objektų išlėkos į aplinkos orą. Sprendžiant ^{137}Cs aktyvumo pažemio ore balanso lygtį parodyta, kad galimos ^{137}Cs emisijos iš veikiančių branduolinės energetikos objektų ir pernašos iš stratosferinio rezervuaro sudaro iki 10% viso radionuklido kiekio pažemio ore. Atgalinės oro masių judėjimo trajektorijos buvo skaičiuojamos šuoliškiems ^{137}Cs aktyvumo koncentracijos ore padidėjimų (>8 μBq m^{-3}) laikotarpiams, naudojant „Hybrid Single Particle Lagrangian Integrated Trajectory" (HYSPLIT) modelį. Parodyta, kad šie ^{137}Cs aktyvumo koncentracijos ore padidėjimai yra susiję su aerozolio, ^{137}Cs nešiklio, pernaša su oro masėmis iš užterštų po Černobylio AE avarijos rajonų.