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ARTIFICIAL RADIONUCLIDES AND THEIR INTERACTION WITH AEROSOL PARTICLES IN THE IGNALINA NPP AND ITS ENVIRONMENT

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VILNIAUS UNIVERSITETAS
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DIRBTINIAI RADIONUKLIDAI IR JŲ SĄVEIKA SU AEROZOLIO DALELĖMIS IGNALINOS AE IR JOS APLINKOJE

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INTRODUCTION

^{137}Cs is one of the most important artificial radionuclides released into the environment during nuclear weapon tests and the Chernobyl nuclear power plant (NPP) accident that determines ionizing radiation doses to the population up till now. Long-term measurements of radionuclide activity concentrations in the ground-level air and deposition enable evaluating geophysical factors that determine the change of radionuclide activity concentrations over a time [1]. These measurements carried out at the geophysical station of the Institute of Physics (PhI station), located in the Ignalina NPP impact zone, enable the investigation of the impact of the Ignalina NPP and other sources of artificial radionuclide emission in the air [2].

The Ignalina NPP RBMK type reactor contains one cooling loop with the branch for the overheated steam, in which the steam condensation takes place in the presence of high ionizing radiation doses. Conditions of radionuclide interaction with aerosol particles (temperature, water steam pressure, chemical composition) on the way from the radionuclide formation place to the ground-level air change in a broad range, which can result in the formation of radioactive aerosol particles due to the absorption of water molecules.

The classical method of the radionuclide activity determination in aerosol particle samples, collected on filters, does not provide information whether aerosol particles are formed due to the absorption of water molecules. The leaching of aerosol particle samples using the distilled water and measuring of radionuclide activities in samples before and after the experiment was studied in order to investigate the presence of radionuclides in soluble and insoluble fractions of aerosol particles.

The objects of this work were artificial radionuclides and their interaction with aerosol particles in the system "Ignalina NPP – environment".

Aims of this work were:

– to determine sources of ^{137}Cs in the Ignalina NPP environment air and evaluate changes of the long-term trend of ^{137}Cs activity concentrations in the Ignalina NPP ground-level air.

– to evaluate the radionuclide distribution in soluble and insoluble fractions of aerosol particles in the system „Ignalina NPP – environment".

In order to achieve these aims, the following **tasks** were established:

– to permanently measure radionuclide activity concentrations in the ground-level air and perform the statistical analysis of experimental data.

– to determine sources of ^{137}Cs transport in the Ignalina NPP environment air during cases of episodic increases in ^{137}Cs activity concentrations in the air.

– to measure artificial radionuclide activity concentrations in deposition and evaluate their flux from the air to the ground surface in 2005-2008.

– to investigate the leaching of aerosol particle samples, collected in the ground-level air and in the Ignalina NPP ventilation system gases, and measure radionuclide activities in samples before and after the experiment.

– to propose the calculation method to estimate the change in the distribution of radionuclides in soluble and insoluble fractions of aerosol particles in the NPP plume.

Novelty of the work:

For the first time the change in mean annual ^{137}Cs activity concentrations in the air in the Ignalina NPP region in 1978-2008 was described using the exponential function, and the half-time of the change in mean annual ^{137}Cs activity concentrations in the air was calculated.

For the first time the growth of aerosol particles, radionuclide carriers, in the cooling water steam in the one contour NPP condenser was investigated.

Statements presented for defence:

1. In recent years annual ^{137}Cs activity concentrations in the ground-level air in the Ignalina NPP region change with the half-time that is close to the ^{137}Cs decay half-time.
2. In the one contour nuclear power plant coolant water steam, noble gas radionuclides decay into alkaline and earth-alkali metal elements, which become condensation nuclei and due to the absorption of water molecules in the oversaturated water vapour grow into radioactive aerosol particles.

Content and structure of the work:

The dissertation is written in Lithuanian. The work consists of the introduction, the literature survey, presentation of methods, investigation results, results, references and an annex. The work comprises 127 pages, 46 figures, 22 tables and 143 references.

METHODS OF THE WORK

The radioecological monitoring of the Ignalina NPP environment has been carried out at the PhI station (N 55°34'03", E 26°35'26"), situated at a 3.5 km distance South-East from the Ignalina NPP (N 55°36'18", E 26°33'36") [3, 4].

For the continuous collection of air-borne radionuclides the air filtration equipment installed at the PhI station of the high airflow rate of about 1800 m^3 h^{-1} through the perchlorvinyl Petrianov filters FPP-15-1.5 was used for aerosol sampling once a week. Filters were pressed into pellets of the standard geometry and activities of radionuclides were measured by the gamma spectrometry method. The ORTEC portable gamma spectrometer with the coaxial HPGe detector in a multilayer shielding was used for the gamma spectrometry. The SNIP (SILENA) multichannel analyzer was used to obtain energy spectra of radionuclides. The radionuclide activity concentrations in the ground-level air were calculated as a ratio between the radionuclide activity in the sample and the volume of the filtered air. The minimum radionuclide detectable activity concentration in the air was 0.1 μBq m^{-3}.

The distribution of radionuclide activity concentrations in the air on a local scale was calculated using semi-empirical Pasquill-Gifford equations and the global model Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) of the admixture dispersion and deposition [5]. Meteorological data obtained from the Ignalina NPP and those of the HYSPLIT meteorological archive were compared and they showed good correlation.

Deposition samples were collected at the PhI station every 1-2 months: rain samples from the 10 m^2 sloping surface into the 80 L bath inside the station, and snow samples from the 1-2 m^2 surface near the PhI station. A method of rapid co-precipitation with metal hydroxides deposits was used to concentrate radionuclides in deposition samples.

The leaching of aerosol particle samples, collected in the ground-level air and in the Ignalina NPP ventilation system gases, was studied by means of sample extraction with distilled water.

RESULTS OF THE RESEARCH

Experimental results of this research include radionuclide activity concentrations in the ground-level air and deposition in the Ignalina NPP region and results on the leaching of aerosol particles collected on filters in the ground-level air and in the Ignalina NPP ventilation system.

1. Radionuclide activity concentrations in the ground-level air

Results of measurements of artificial ^{137}Cs, ^{60}Co, ^{54}Mn radionuclides and cosmogenic ^{7}Be activity concentrations in the Ignalina NPP ground-level air in 2004-2009 are presented and discussed. ^{137}Cs and ^{7}Be activity concentrations in the air are given in Fig. 1.

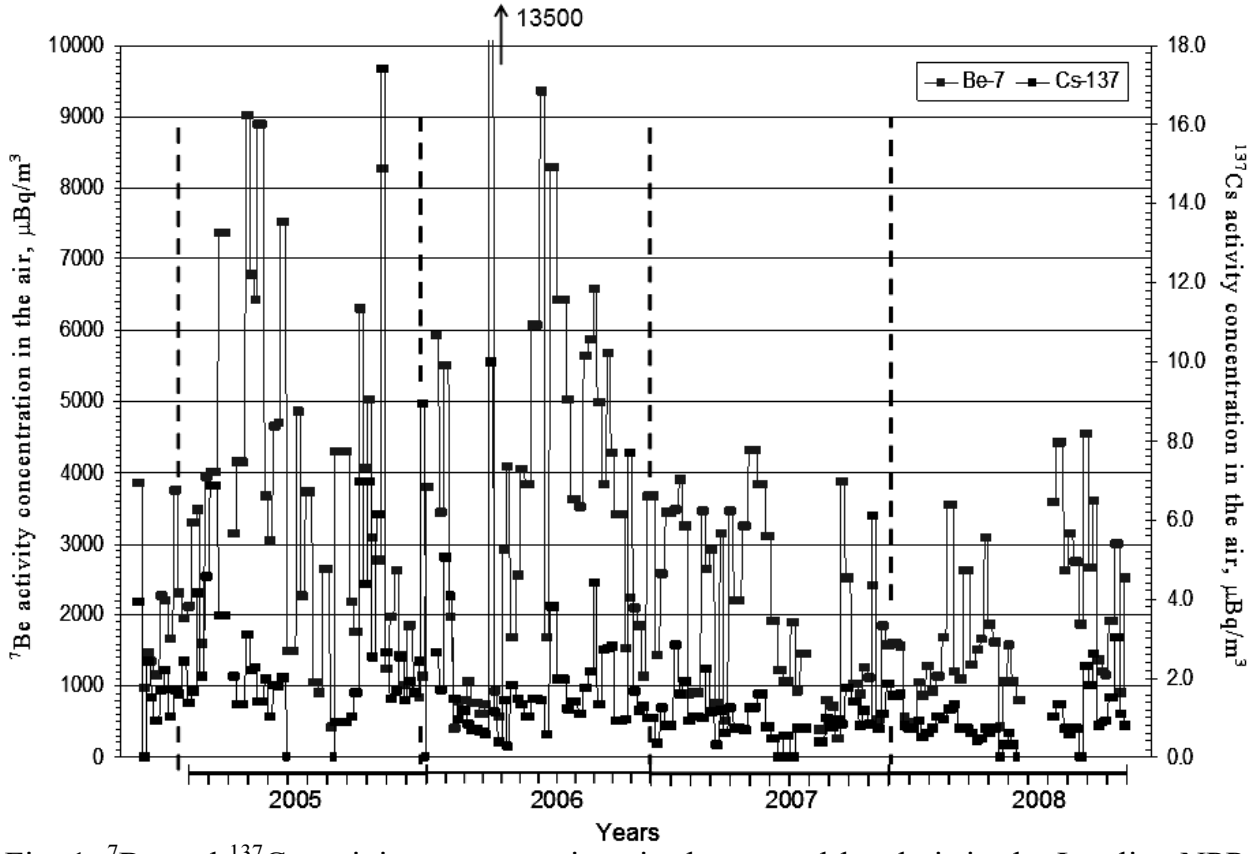

Fig. 1. ^{7}Be and ^{137}Cs activity concentrations in the ground-level air in the Ignalina NPP region on 2004.10.15-2009.01.02.

Seasonal increases in ^{7}Be activity concentrations in the air were observed in spring in 2005-2006. The increase in ^{7}Be activity concentrations in the air was observed in autumn in 2008 and could be explained by changes in the atmosphere circulation in recent years.

^{137}Cs gamma radiation was registered in almost all ground-level air aerosol particle samples. Maximum ^{137}Cs activity concentrations were observed in January-February and September-October. Every year a number of episodic increases in ^{137}Cs activity concentrations in the air which does not coincide with the registration of Ignalina NPP produced ^{60}Co and ^{54}Mn gamma radiation in samples was observed. The number of ^{60}Co

and ^{54}Mn radiation registration cases in samples was decreasing each year, which is explained by the shut-down of the Ignalina NPP Unit 1 in 2004.

Values of weekly radionuclide activity concentrations in the air over a longer time period fluctuate due to a number of factors and can be statistically evaluated by calculating the distribution of radionuclide activity concentrations in the air. Mean annual arithmetic and most probable ^{137}Cs activity concentrations in the air were calculated on the basis of distributions of ^{137}Cs activity concentrations in the air for a period of 2005-2008 (Table 1).

Table 1. Mean annual arithmetic and most probable ^{137}Cs activity concentrations in the air in the Ignalina NPP region in 2005-2008.

Year	^{137}Cs activity concentration in the air, μBq/m^3	
	Mean arithmetic X	Most probable X_C
2005	2.1 ± 1.8	1.4 ± 1.1
2006	1.9 ± 1.8	1.1 ± 1.0
2007	1.1 ± 0.5	0.8 ± 0.9
2008	0.9 ± 0.5	0.6 ± 0.4

These results are used to set up and analyze the trend of mean annual ^{137}Cs activity concentrations in the ground-level air in the Ignalina NPP region in 1978-2008.

1.1 ^{137}Cs activity concentrations in the Ignalina NPP region ground-level air in 1978-2008

The trend of mean annual ^{137}Cs activity concentrations in the ground-level air in 1978-2008 was approximated using the exponential function as follows:

$$C_t = C_o \exp[-\frac{\ln 2(t - t_0)}{T_{1/2}}], \qquad (1)$$

where C_t is the ^{137}Cs activity concentration in the air (μBq m^{-3}) in a particular year t, C_0 is the ^{137}Cs activity concentration in the air (μBq m^{-3}) at the initial moment of time t_0 (year), and $T_{1/2}$ is the half-time of the change in ^{137}Cs activity concentrations in the ground-level air (y). Results of the approximation are given in Fig. 2.

Three periods with a different mean annual radiocaesium activity concentration trend indicating different ^{137}Cs sources in the air can be distinguished. 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 half-time of the change in radionuclide activity concentrations in the ground-level air during 1981-1985 ($T_{1/2}$=9.7 months) was calculated, which evidently shows the global fallout of radionuclides from the stratospheric reservoir to be the main source of ^{137}Cs to the ground-level air till 1986.

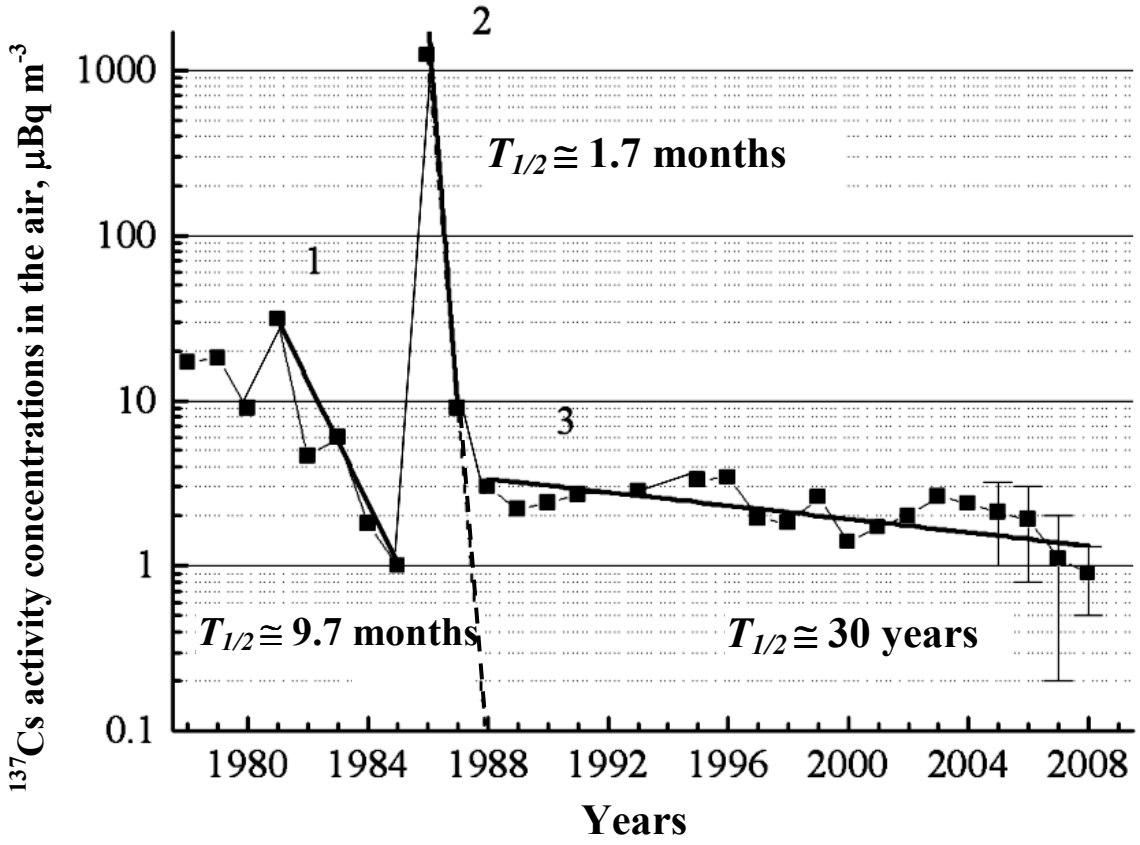

Fig. 2. The trend of mean annual ^{137}Cs activity concentrations in the ground-level air in the Ignalina NPP region in 1978-2008.

The ^{137}Cs emission to the atmosphere during the accident at the Chernobyl 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 change in ^{137}Cs activity concentrations in the ground-level air in 1986-1988 ($T_{1/2}$=1.7 months) indicates the radionuclide fallout from the troposphere.

Results show that since 1988 ^{137}Cs activity concentrations in the air have decreased much slower ($T_{1/2} \cong$ 30 years) than in previous periods. Nowadays mean annual ^{137}Cs activity concentrations in the air decrease with the half-time, which coincides with the ^{137}Cs decay half-time. In recent years ^{137}Cs activity concentrations in the ground-level air almost do no change, because their decrease due to wash-out with precipitation and radioactive decay is compensated by ^{137}Cs emissions into the atmosphere from its sources in the environment.

1.2 Analysis of episodic increases in ^{137}Cs activity concentrations in the air

Episodic increases in weekly ^{137}Cs activity concentrations during aerosol sampling periods that do not coincide with cases of registration of Ignalina NPP produced radionuclides in samples were registered at the PhI station. 8 cases of episodic increases in weekly ^{137}Cs activity concentrations in 2004-2008 were analyzed using the method of calculation of air mass transport backward trajectories, a part of the HYSPLIT model. Backward trajectories of air mass transport to the Ignalina NPP region that coincide with

the aerosol sampling period on October 30 – November 5, 2005 at the PhI station are given in Fig. 3.

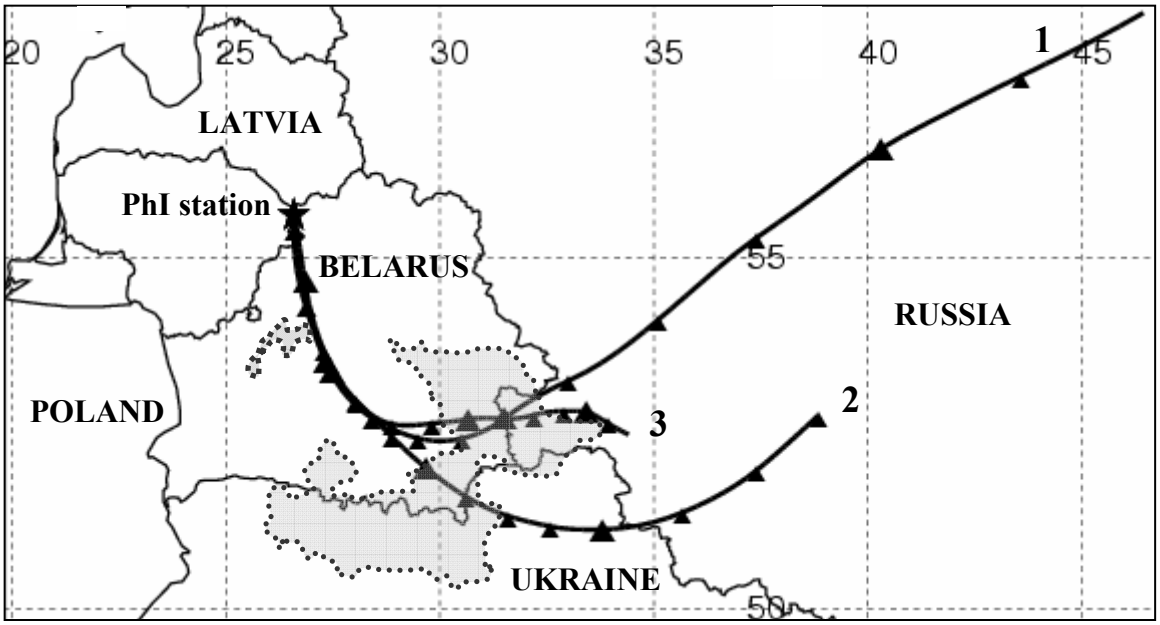

Fig. 3. Backward trajectories of air mass transport to the Ignalina NPP region that coincide with the aerosol sampling period on October 30 – November 5, 2005 at the PhI station. 1 – the air mass transport backward trajectory ending on October 30, 2005 12:00 at the PhI station, 2 – ending on November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

The results show that in almost all cases episodic increases in ^{137}Cs activity concentrations in the air correlate with air mass transport trajectories that intersect territories, polluted with ^{137}Cs after the accident at the Chernobyl NPP.

1.3 Radionuclide activity concentrations in deposition

Measurement results of radionuclide activity concentrations in deposition in the Ignalina NPP region in 2005-2008 are given Table 2.

Table 2. The radionuclide activity concentrations in deposition in 2005-2008.

Sampling period	Radionuclide flux from the air to the ground surface, F, Bq/(m^2 month)					
	Fine particle fraction			Coarse particle fraction		
	^{137}Cs	^{60}Co	^{7}Be	^{137}Cs	^{60}Co	^{7}Be
1	2	3	4	5	6	7
2005.01.23-03.17	0.5	0.11	28	0.021	0.006	0.25
2005.04.20-05.29	1.5	< 0.03	22	0.012	< 0.002	0.14
2005.05.29-07.12	1.5	0.12	26	0.011	< 0.002	< 0.05
2005.07.12-09.26	1.7	0.10	19	0.005	0.03	< 0.05
2005.09.26-11.05	0.8	< 0.03	1,8	0.011	< 0.002	< 0.05
2006.01.31-03.12	0.9	0.06	41	0.020	0.011	< 0.05
2006.03.12-07.20	1.2	0.06	17	0.012	0.005	< 0.05

1	2	3	4	5	6	7
2006.07.20-25	2.2	< 0.03	21	0.109	0.042	< 0.05
2006.07.25-08.07	1.8	< 0.03	16	0.072	0.016	< 0.05
2006.08.07-09.27	1.6	0.10	11	0.009	< 0.002	< 0.05
2006.09.27-11.03	0.5	0.05	9	0.022	< 0.002	< 0.05
2007.01.20-03.01	0.9	0.08	21	0.023	0.012	0.34
2007.03.01-06.06	0.9	0.05	19	0.015	< 0.002	0.11
2007.06.06-07.10	1.0	< 0.03	23	0.009	< 0.002	< 0.05
2007.07.10-10.06	0.7	< 0.03	10	0.008	< 0.002	< 0.05
2007.10.06-11.17	1.1	0.07	8	0.017	< 0.002	< 0.05
2008.01.31-03.06	0.8	< 0.03	25	0.018	< 0.002	0.18
2008.03.06-04.23	1.8	< 0.03	23	0.020	< 0.002	< 0.05
Mean value:	**1.1 ± 0.3**	**0.05 ± 0.02**	**18 ± 3**	**0.015 ± 0.005**	**0.005 ± 0.002**	**0.05 ± 0.01**

Experimental results enable evaluating the flux of radionuclides into the ground-level air from all radionuclide sources in the environment using a two-box model. Calculation results show that the ^{137}Cs emission from the Ignalina NPP contribute up to 10 % of the radionuclide flux to the Ignalina NPP region.

1.4 Short-lived radionuclide activity concentrations in the INPP ventilation air

Radionuclide activity concentrations in the Ignalina NPP ventilation system air were measured in order to determine activity concentrations of noble gases decay products in the air at intermediate sampling points on the radionuclide path to the ventilation stack (Table 3).

Table 3. Radionuclide activity concentrations in the Ignalina NPP ventilation system air.

Radionuclide	Delay chamber		Activity reducing unit	
	Before the chamber	Behind the chamber	Before the unit	Behind the unit
^{88}Rb	4.5×10^{10}	3.2×10^{8}	3.6×10^{7}	–
^{91}Sr	7.4×10^{7}	1.1×10^{7}	–	7.8×10^{3}
^{131}I	4.7×10^{6}	4.9×10^{5}	1.1×10^{6}	4.2×10^{2}
^{137}Cs	1.3×10^{7}	2.6×10^{6}	3.4×10^{3}	6.3×10^{2}
^{138}Cs	2.5×10^{10}	2.3×10^{9}	3.4×10^{6}	1.7×10^{6}
^{139}Ba	1.6×10^{10}	5.8×10^{8}	2.0×10^{6}	1.5×10^{6}
^{140}Ba	3.4×10^{7}	7.6×10^{6}	2.1×10^{4}	–

Activity concentrations of ^{88}Rb, ^{138}Cs and ^{139}Ba, collected on aerosol filters, in the turbine ejector gas were found to be in the interval of 3.4×10^{7} – 4.5×10^{10} Bq/m^3. These results can be explained by the fact that noble gas radionuclides overcome the water and steam boundary in drum-separators, pass through the turbine condenser and decay to alkaline and earth-alkali elements that become aerosol formation centres, absorb water molecules and in less than several minutes grow into aerosol particles, collected on aerosol filters. This process is similar to the decay of emanation (^{222}Rn, ^{220}Tn, ^{219}An) radionuclides and formation of daughter nuclei that are rapidly attached to aerosol particles.

2. Leaching of aerosol particle samples collected on filters

The leaching of aerosol particles was investigated in samples collected on filters in different places of the system „Ignalina NPP – environment“: 1) in the ground-level air in the Ignalina NPP region in 2004-2005 when PhI station was on the lee side of the Ignalina NPP, 2) in the ventilation stack of the Ignalina NPP operating reactor and the shut-down reactor in 2006-2007 and additionally in the ventilation system of the operating reactor in 2008.

2.1 Leaching of aerosol particles collected on filters in the ground-level air

Results on the leaching of aerosol particles collected on filters in the ground-level air in the Ignalina NPP region in 2004-2005 are given in Table 4.

Table 4. ^{137}Cs and ^{60}Co activities in aerosol particle samples before the leaching experiment and after the experiment and the ratio between the radionuclide activity in the leached sample and in the initial sample, W, %

Nr	Sampling period	^{137}Cs			^{60}Co		
		Activity, mBq		W, %	Activity, mBq		W, %
		A	A_{nt}		A	A_{nt}	
2906	2004.03.20-30	440	270	39 ± 10	165	130	21 ± 7
2922	2004.07.26-08.03	305	210	31 ± 8	300	230	23 ± 6
2947	2005.01.02-08	170	120	29 ± 7	270	210	22 ± 7
2948	2005.01.29-02.05	1250	390	69 ± 15	105	80	24 ± 6
2956	2005.04.21-05.01	495	420	15 ± 3	760	570	25 ± 6
2985	2005.12.27-31	420	370	12 ± 3	190	140	26 ± 7
	Mean value:			–			**24 ± 7**
2973	2005.09.26-10.02	1640	1170	29 ± 6	–	–	–
2974	2005.10.02-09	1230	810	34 ± 7	–	–	–
2977	2005.10.22-30	860	490	43 ± 7	–	–	–
2978	2005.10.30-11.05	4280	2215	48 ± 7	–	–	–
2982	2005.11.26-12.04	185	130	30 ± 5	–	–	–

The mean ratio between ^{137}Cs activities in leached samples after the experiment and in initial samples, collected in the ground level air, was 25 %, and that of ^{60}Co was 24 %. These values are close to the mean mass content of soluble aerosol particles in the continental air mass – 28% [6]. The mean quantity of ^{137}Cs and ^{60}Co in the soluble fraction of aerosol particles, collected in the ground-level air, was 32 % and 22 %, respectively [7].

The ratio between ^{137}Cs activities in leached samples after the experiment and in initial samples, collected in cases of air mass transport from Chernobyl NPP accident polluted regions, was in the interval of 43-69 %. The quantity of ^{137}Cs in the soluble fraction of aerosol particles, collected in the ground-level air after forest fires in the Chernobyl NPP polluted regions, was up to 60 % [8].

Experimental results on the leaching of aerosol particles correlate well with the published data on the radionuclide distribution in soluble and insoluble fractions of aerosol particles.

2.2 Leaching of aerosol particles, collected on filters in the Ignalina NPP ventilation system

Results on the leaching of aerosol particles, radionuclide carriers, collected on filters in the ventilation stack of the Ignalina NPP operating reactor and the shut-down reactor in 2006-2007 are given in Table 5.

Table 5. ^{137}Cs and ^{60}Co activities in aerosol particle samples before the leaching experiment and after the experiment and the ratio between the radionuclide activity in the leached sample after the experiment and in the initial sample, *W*, %.

Sampling period	^{137}Cs			^{134}Cs			^{54}Mn			^{60}Co		
	Activity, mBq		*W*, %	Activity, mBq		*W*, %	Activity, mBq		*W*, %	Activity, mBq		*W*, %
Unit 2 operating reactor	*A*	A_{nt}		*A*	A_{nt}		*A*	A_{nt}		*A*	A_{nt}	
2006.01.10-19	190	65	66±6	–	–	–	130	45	65±15	420	120	71±17
2007.06 01-07	1760	210	88±7	85	25	71±25	90	40	56±20	195	70	64±14
2007.06.07-15	2040	505	75±6	125	40	68±20	165	50	70±15	270	80	70±16
2007.06.19-26	1580	265	83±10	105	45	57±15	170	35	79±20	605	190	69±16
2007.06.26-07.03	350	120	66±6	60	–	–	1030	300	71±17	2420	830	66±15
2007.07.10-16	665	180	73±8	70	70	0	225	60	73±20	470	150	68±17
2007.07.17-24	165	50	70±7	–	–	–	225	–	–	310	120	61±14
Mean value			**74 ± 22**			**–**			**69 ± 25**			**67 ± 17**
Unit 1 shut-down reactor												
2005.12.23-2006.01.03	215	160	26±3	–	–	–	–	–	–	185	145	22±3
2006.01.3-9	235	170	28±3	–	–	–	–	–	–	210	160	24±4
2006.01.23-29	60	50	17±2	–	–	–	30	–	–	150	130	13±2
2007.06 01-07	40	35	12±2	–	–	–	–	–	–	155	110	29±5
2007.06.07-15	120	100	17±2	–	–	–	40	–	–	250	220	12±3
2007.06.19-26	295	230	22±3	–	–	–	–	–	–	250	200	20±4
2007.06.26-07.03	780	485	38±5	605	360	40±10	35	–	–	345	290	16±3
2007.07.10-16	60	40	33±4	40	–	–	–	–	–	240	190	21±4
Mean value			**24 ± 5**			**–**			**–**			**20 ± 5**

The significant difference of ratios between ^{137}Cs activities in leached samples after the experiment and in initial samples, collected in the Ignalina NPP operating reactor and the shut-down reactor ventilation system, was determined: 74 ± 22 % and 24 ± 5 %, respectively, and that of ^{60}Co was 67 ± 17 % and 20 ± 5 %, respectively.

The quantity of ^{137}Cs and ^{60}Co in the leached fraction of aerosol particles, collected in the shut-down reactor ventilation air, correlates with the mean mass content of soluble aerosol particles in the continental air mass – 28% [6]. Results of the leaching of aerosol particles, collected in the operating reactor ventilation air, are close to values of quantity

of ^{137}Cs and ^{60}Co in the soluble fraction of aerosol particles, collected in the NPP reactor ventilation air – 78 % and 99 %, respectively [7].

The radionuclide activity ratios in leached samples and in initial samples, probably, reflect the ratio between the quantity of radionuclides, associated with aerosol particles that mostly consist of water molecules, and radionuclides in the insoluble fraction of aerosol particles, that remain on the filter.

The water evaporation and steam condensation in the presence of the oversaturated water vapour pressure take place only in the operating NPP reactor. Conditions of the soluble aerosol particle formation are favourable in ejector gases from the turbine that mainly consist of water vapour. This process is very close to the investigated formation of aerosol particles in the presence of saturated water vapour [9, 10].

In the leaching experiment aerosol particles of stable structures (due to surface tension forces), made of water molecules, are ruined. The thermal movement of water molecules in the solution destroys the arrangement of water molecules on the surface of aerosol particles that results in the transfer of water molecules and radionuclides into the solution. In literature aerosol particles, formed due to the absorption of water molecules, are entitled as the soluble aerosol particle fraction. Apparently, experimental results of this work are close to results on the aerosol particle growth under conditions of saturated vapour as described in literature.

The leaching of aerosol particles, collected in components of the ventilation system of the operating Ignalina NPP reactor, was investigated (Table 6).

Table 6. The ratio between ^{137}Cs and ^{60}Co activities in leached samples after the experiment and in initial samples, *W*, %.

Sampling point	*W*, %,	
	^{137}Cs	^{60}Co
Delay chamber inlet	67 ± 5	–
Delay chamber outlet	80 ± 5	50 ± 10
Activity reducing unit inlet	63 ± 5	60 ± 10
Activity reducing unit outlet	71 ± 5	67 ± 15

These results are close to leaching results of aerosol particles, collected in the ventilation stack of the operating Ignalina NPP reactor. For illustration purposes results on the leaching of aerosol particles, ^{60}Co carriers, collected on filters in the ventilation system of the operating reactor, in the ventilation stack of the shut-down reactor and in the ground level air, are plotted in Fig. 4.

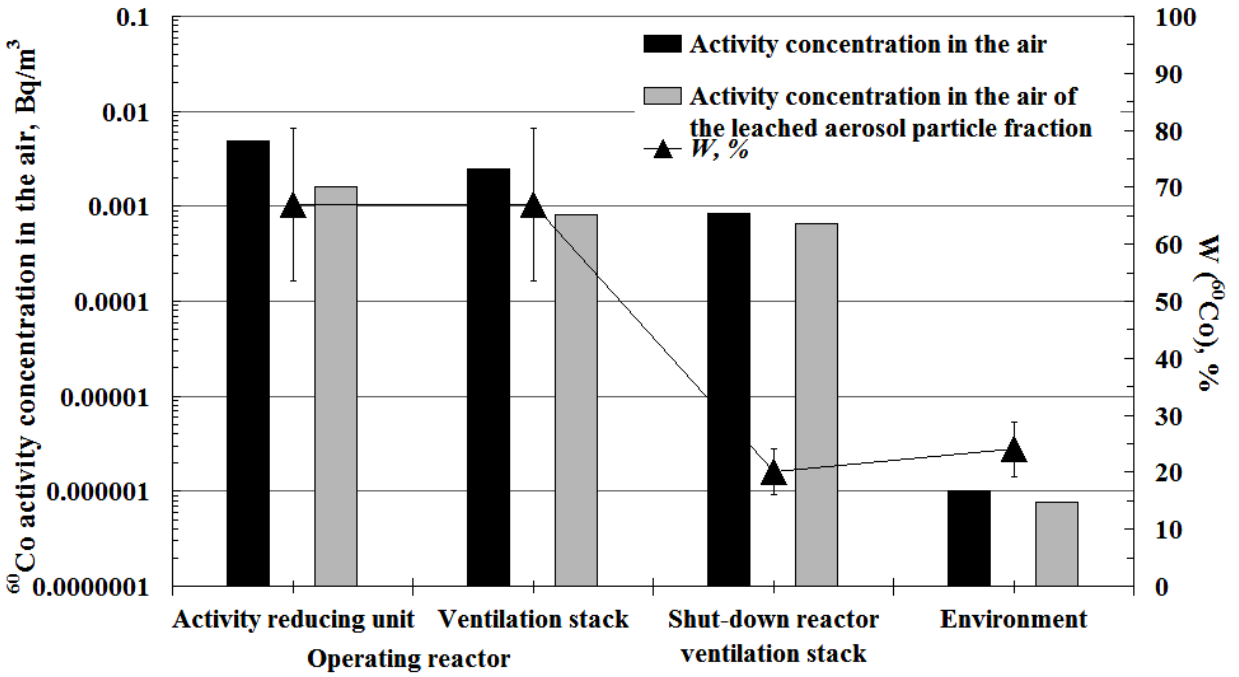

Fig. 4. The ^{60}Co activity concentration and the activity concentration of the leached fraction of aerosol particles, ^{60}Co carriers, in the air of the ventilation system of the operating Ignalina NPP reactor, in the ventilation stack of the shut-down Ignalina NPP reactor and in the environment (bars, left axis) and the mean ratio between the ^{60}Co activity in the leached sample and the initial sample, *W* % (points and line, right axis).

2.3 Transport of soluble and insoluble aerosol particles in the ground-level air

The change in the leaching of aerosol particles, ^{60}Co carriers, collected in the ventilation stack of the operating reactor and in the ground-level air, can be estimated by supplementing the semi-empirical Pasquill-Gifford equation with the coefficient of soluble ^{60}Co aerosol particle scavenging, F_R:

$$F_R = \exp - \frac{k_t x}{u}, \qquad (2)$$

where k_t is the rate of the change in the leaching of aerosol particles, ^{60}Co carrier, at a distance x from the emission source, s^{-1}, u is the wind velocity, m s^{-1}, x is the distance between the stack and the sampling point of the ground-level air aerosol particles, m.

The example of calculation of the radionuclide activity concentration in the air, *C (x, y, z)*, at the measurement point: 1) with no regard to the distribution of ^{60}Co in the soluble aerosol particle fraction (coefficient F_R is not estimated) and 2) with regard to the distribution of ^{60}Co in the soluble aerosol particle fraction (coefficient F_R is estimated), is given in Fig. 5.

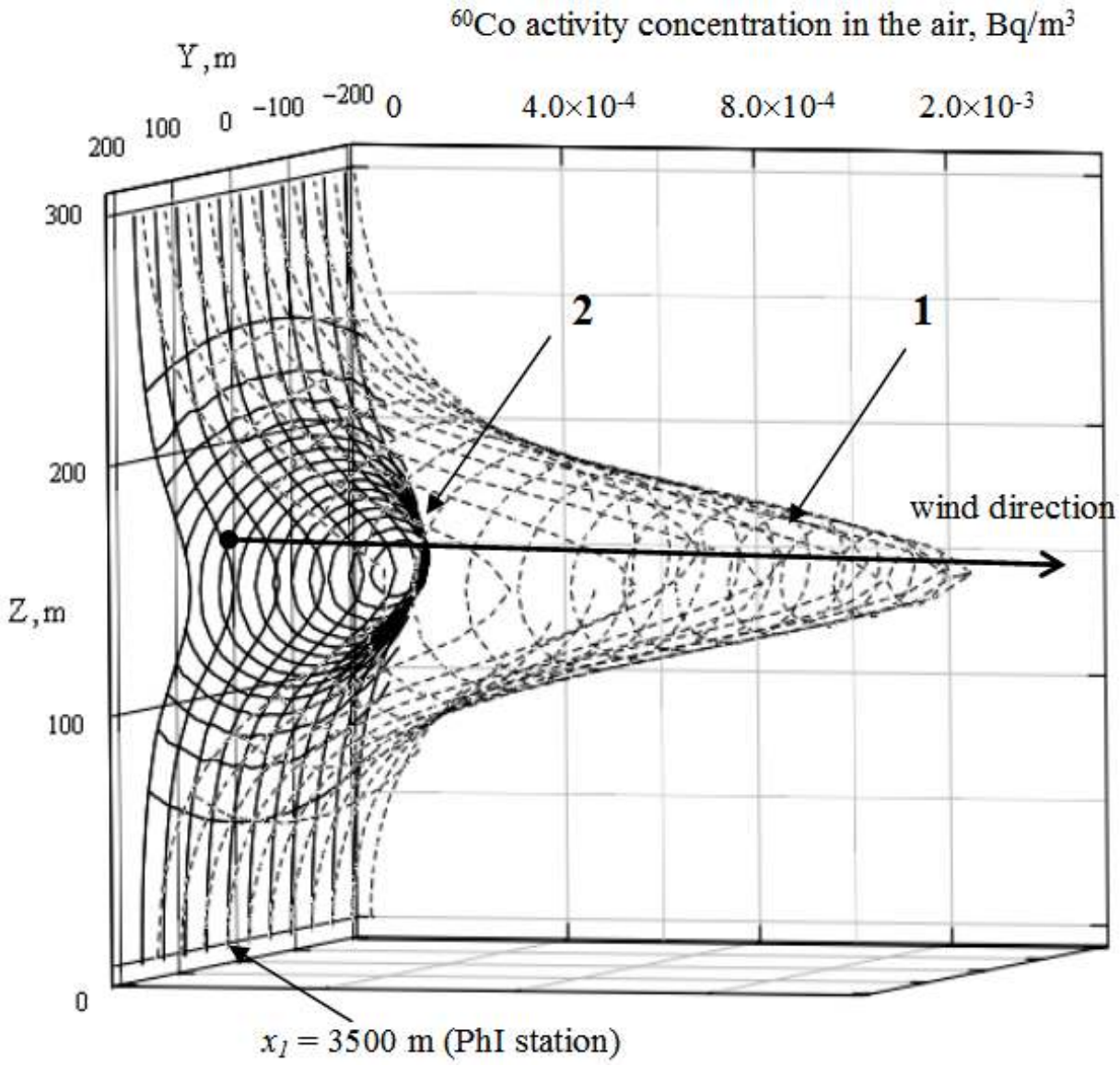

Fig. 5. The distribution of the ^{60}Co activity concentration in the air at the *y,z* plane at the PhI station at the distance *x*=3500 m from the Ignalina NPP: 1 – with no regard to the distribution of ^{60}Co in the soluble aerosol particle fraction (coefficient F_R is not estimated) and 2 – with regard to the distribution of ^{60}Co in the soluble aerosol particle fraction (coefficient F_R is estimated).

The method of the distribution of the mixture of aerosol particles, radionuclide carriers, that is characterized by the altering in time and space ratio of the ^{60}Co activity concentration in the air and the ^{60}Co activity concentration in the air in the leached aerosol particle fraction, calculated by supplementing the semi-empirical Pasquill-Gifford equation with the coefficient of soluble ^{60}Co aerosol particle scavenging was introduced.

CONCLUSIONS

1. The trend of mean annual ^{137}Cs activity concentrations in the ground-level air in the Ignalina NPP region was described using the exponential function with the 30 year half-time of the change in ^{137}Cs activity concentrations in the ground-level air.
2. The mean ^{137}Cs activity concentration in the deposition in the Ignalina NPP region in 2005-2008 was calculated to be 1.1 Bq/(m^2 month).
3. Episodic increases in ^{137}Cs activity concentrations in the air correlate well with air mass transport trajectories that intersect territories, polluted with ^{137}Cs after the accident at the Chernobyl NPP.
4. In the one contour nuclear power plant coolant water steam, noble gas radionuclides decay into alkaline and earth-alkali metals, which become condensation nuclei and due to the absorption of water molecules grow into radioactive aerosol particles.
5. Quantity of ^{137}Cs and ^{60}Co in the leached part of aerosol particles, collected in the ventilation air of the operating reactor, was 74 ± 22 % and 67 ± 17 %, respectively, and in the ventilation air of the shut-down reactor was 24 ± 5 % and 20 ± 5 %.
6. The distribution of the mixture of aerosol particles, radionuclide carriers, that is characterized by the altering in time and space ratio of the ^{60}Co activity concentration in the air and ^{60}Co activity concentration in the air in the leached aerosol particle fraction, was calculated by supplementing the semi-empirical Pasquill-Gifford equation with the coefficient of soluble ^{60}Co aerosol particle scavenging.

REFERENCES

[1] B. Styra. Branduolinės meteorologijos klausimai. Vilnius: LTSR Mokslų Akademija, 418 p. (1959).
[2] V. Valkovic. Radioactivity in the Environment. Elsevier, 681 p. (2000).
[3] Р. Ясюленис, Р. Кренявичюс, Л. Милошене, А. Серапинас. О распределении концентраций ^{137}Cs и ^{7}Be в атмосферном воздухе в районе строящейся Игналинской АЭС. Проблемы исследования загрязнения атмосферы. Физика атмосферы 7, 61-65 (1981).
[4] R. Jasiulionis. Radionuklidų srautų Ignalinos AE aplinkoje įvertinimas. Lietuvos valstybinės mokslo programos "Atominė energetika ir aplinka" mokslinių ataskaitų rinkinys. Vilnius, 1-40 (1997).
[5] R. Draxler, G. Hess. An overview of the HYSPLIT4 modelling system for trajectories, dispersion and deposition. Australian Meteorology Magazine 47 (http://www.arl.noaa.gov/HYSPLIT.php), 295-308 (1998).
[6] B. Svenningsson, H. Hansson, A. Wiedensohler, K Noone, J. Ogren, A. Hallberg, R. Colvile. Hygroscopic growth of aerosol particles and its influence on nucleation scavenging in cloud: Experimental results from Kleiner Feldberg. Journal of Atmospheric Chemistry 19 (1-2), 129-152 (1994).
[7] R. Jasiulionis, I. Savickaitė. Radionuclides in ground-level air and deposits near the Ignalina NPP. Nukleonika 46 (4), 183-187 (2001).
[8] G. Lujanienė, V. Lujanas, A. Mikelinksienė. Forest fires as possible mechanism of redistribution of radioactive contamination. Sveikatos mokslai 3, 11-16 (2003).

[9] M. Kulmala, L. Pirjola and J. Mäkelä. Stable sulphate clusters as a source of new atmospheric particles. Nature 404, 66-69 (2000).
[10] M. Kulmala et al. The condensation particle counter battery (CPCB): a new tool to investigate the activation properties of nanoparticles. Journal of Aerosol Science 38 (3), 289-304 (2006).

LIST OF PUBLICATIONS

1. R. Jasiulionis, A. Rožkov and L. Vyčinas. Radionuclides in the ground-level air and deposition in the Ignalina NPP region during 2002-2005. Lithuanian Journal of Physics 46 (1), 101-108 (2006).
2. R. Jasiulionis, A. Rožkov. Radionuclides in deposition in the Ignalina NPP region in 2005. Central European Journal of Physics 4 (4), 417-428 (2006).
3. R. Jasiulionis and A. Rožkov. ^{137}Cs activity concentration in the ground-level air in the Ignalina NPP region. Lithuanian Journal of Physics 47 (2), 195-202 (2007).
4. R. Jasiulionis, A. Rožkov. The solubility of the aerosol, ^{137}Cs and ^{60}Co carrier, in the Ignalina NPP region. Applied Radiation and Isotopes 66 (12), 1992-1998 (2008).
5. R. Jasiulionis, L. Vyčinas, A. Rožkov. In situ study of the ^{41}Ar plume released from the Ignalina NPP. Central European Journal of Physics 6 (4), 775-780 (2008).
6. R. Jasiulionis, A. Rožkov. Radionuclides in deposition in the Ignalina NPP region in 2005. Proceedings of the Conference „Metals in the Environment“, 53-60, (2006).
7. R. Jasiulionis and A. Rožkov. ^{137}Cs in the ground-level air and deposition in the Ignalina Nuclear Power Plant region. American Institute of Physics Conference Procedings 889, 417-418 (2007).

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DIRBTINIAI RADIONUKLIDAI IR JŲ SĄVEIKA SU AEROZOLIO DALELĖMIS IGNALINOS AE IR JOS APLINKOJE

Reziumė

Disertaciją sudaro įvadas, spausdintų darbų sąrašas, literatūros apžvalga, metodikos aprašymas, rezultatai, cituotos literatūros sąrašas ir priedas. Darbo apimtis 127 puslapiai teksto, 46 paveikslėliai, 22 lentelės ir 143 cituoti literatūros šaltiniai.

Darbo tyrimo objektas yra dirbtiniai radionuklidai ir jų sąveika su aerozolio dalelėmis Ignalinos AE ir jos aplinkoje. Darbe nustatyti ^{137}Cs patekimo į IAE aplinkos orą šaltiniai ir įvertinti ilgamečiai ^{137}Cs aktyvumo koncentracijų pokyčiai. Įvertintas dirbtinių radionuklidų pasiskirstymas tirpioje ir netirpioje aerozolio dalelių frakcijose sistemoje „IAE – aplinka“.

Problemos aktualumas. Radionuklidų patekimas į aplinką yra aktuali problema visose branduolinės energetikos ciklo grandyse ir visose atominių elektrinių paleidimo, darbo ir eksploatacijos nutraukimo stadijose. ^{137}Cs yra vienas svarbiausių dirbtinių radionuklidų, patekusių į aplinką branduolinio ginklo bandymų metu ir įvykus avarijai Černobylio AE, iki šiol lemiantis jonizuojančiosios spinduliuotės apšvitos dozes.

Radionuklidų aktyvumo koncentracijų ore ir iškritose matavimai nuo 2004 iki 2009 metų buvo vykdomi Fizikos instituto geofizinėje stotyje (FI stotis), esančioje Ignalinos AE poveikio zonoje. Tokie ilgalaikiai matavimai leidžia įvertinti geofizinius faktorius, kurie lemia aktyvumo koncentracijų kitimą laiko bėgyje ir įgalina nustatyti Ignalinos AE ir kitų dirbtinių radionuklidų emisijos į orą šaltinių poveikį. Gauta, kad metinės ^{137}Cs aktyvumo koncentracijos pažemio ore pastaraisiais metais kinta su mažėjimo pusiau trukme, kuri artima ^{137}Cs skilimo pusamžiui.

Ignalinos AE RBMK reaktorius turi vieno kontūro aušinimo sistemą su atšaka perkaitintam garui, kurioje vyksta garo kondensacija esant didelėms jonizuojančiosios spinduliuotės dozėms. Aerozolio dalelių ir radionuklidų sąveikos sąlygos (temperatūra, vandens garų slėgis, cheminė sudėtis) visame kelyje nuo radionuklidų pasigaminimo iki pažemio oro keičiasi plačiose ribose ir susidaro sąlygos radioaktyvių aerozolio dalelių augimui dėl vandens molekulių absorbcijos. Nustatytas radioaktyvių aerozolio dalelių augimas prisotintuose vandens garuose veikiančio RBMK reaktoriaus kondensatoriuje. Vieno kontūro atominės elektrinės aušinamame vandens gare inertinių dujų radionuklidai, skildami virsta šarminių ir žemės šarminių metalų elementais, tampa aerozolio gamybos centrais, absorbuoja vandens molekules ir per keliolika minučių ant jų, kaip kondensacijos branduolių, išauga radioaktyvios aerozolio dalelės.

Klasikinis radionuklidų aktyvumo aerozolio dalelių, surinktų filtrais, bandiniuose nustatymas papildytas išplovimo distiliuotu vandeniu eksperimentais, matuojant radionuklidų aktyvumą bandiniuose iki ir po eksperimento. Eksperimente vyksta labiausiai tikėtinas aerozolio dalelių, susidarančių dėl vandens molekulių absorbcijos, sugriovimo procesas ir bandiniuose po eksperimento pasilieka radionuklidai buvę netirpioje aerozolių frakcijoje, o buvę tirpioje frakcijoje pereina į tirpalą. Gauta, kad ^{137}Cs ir ^{60}Co dalis aerozolio dalelių, surinktų veikiančio reaktoriaus ventiliaciniame ore, tirpioje frakcijoje buvo 74 ± 22 % ir 67 ± 17 %, atitinkamai. Aerozolio dalelių, surinktų neveikiančio reaktoriaus ventiliaciniame ore, tirpioje frakcijoje ^{137}Cs ir ^{60}Co dalis buvo 24 ± 5 % ir 20 ± 5 %, atitinkamai.

Naujumas: Pirmą kartą eksponentine funkcija aprašyta metinių ^{137}Cs aktyvumo koncentracijų ore kaita Ignalinos AE regione 1978-2008 metais ir įvertinta ^{137}Cs aktyvumo koncentracijų ore mažėjimo pusiau trukmė. Pirmą kartą nustatytas tirpių aerozolio dalelių, radionuklidų nešėjų, augimas aušinimo vandens gare vieno kontūro atominės elektrinės kondensatoriuje.

Ginamieji teiginiai:

1. Metinės ^{137}Cs aktyvumo koncentracijos pažemio ore pastaraisiais metais kinta su mažėjimo pusiau trukme, kuri artima ^{137}Cs skilimo pusamžiui.
2. Inertinių dujų radionuklidai, patekę į vieno kontūro atominės elektrinės aušinimo vandens garą, skildami virsta šarminių ir žemės šarminių metalų elementais, tampa kondensacijos branduoliais ir dėl vandens molekulių absorbcijos persotintuose vandens garuose auga radioaktyvios aerozolio dalelės.

Darbo rezultatai:

1. Metinių ^{137}Cs aktyvumo koncentracijų ore kaita regione 1988-2008 m. aprašyta eksponentine funkcija su mažėjimo pusiau trukme $T_{1/2} \cong 30$ metų.
2. Vidutinė ^{137}Cs aktyvumo koncentracijos iškritose Ignalinos AE aplinkoje 2005-2008 m. reikšmė gauta lygi 1,1 Bq/(m^2 mėnuo).
3. Šuoliški ^{137}Cs aktyvumo koncentracijų pažemio ore padidėjimai koreliuoja su oro masių pernašos trajektorijomis, kertančiomis teritorijas, užterštas po avarijos Černobylio AE iki šiol liekančias ^{137}Cs patekimo į orą šaltiniu.
4. Vieno kontūro atominės elektrinės aušinamame vandens gare inertinių dujų radionuklidai skildami virsta šarminių ir žemės šarminių metalų elementais, tampa aerozolio gamybos centrais, absorbuoja vandens molekules ir per keliolika minučių ant jų, kaip kondensacijos branduolių, išauga radioaktyvios aerozolio dalelės.
5. ^{137}Cs ir ^{60}Co dalis išplautoje aerozolio dalelių, surinktų veikiančio Ignalinos AE reaktoriaus ventiliaciniame ore, frakcijoje buvo 74 ± 22 % ir 67 ± 17 %, atitinkamai, o neveikiančio reaktoriaus ventiliaciniame ore – 24 ± 5 % ir 20 ± 5 %, atitinkamai.
6. ^{60}Co aktyvumo koncentracijos ore ir išplautoje aerozolio dalelių frakcijoje su kintančiu laike ir erdvėje santykiu pasiskirstymas AE fakele apskaičiuotas, papildant pusiau empirinę Pasquill-Gifford lygtį koeficientu, aprašančiu tirpių aerozolio dalelių, ^{60}Co nešėjų, pašalinimą.

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