Radionuclides in ground-level air and deposits near the Ignalina NPP

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Abstract Results of systematic measurements of radionuclide activity concentrations in the air and deposits from January 1997 to July 2000 near the Ignalina Nuclear Power Plant (hereafter detoted as NPP) are presented. The data on NPP produced radionuclides ^{60}Co and ^{54}Mn in deposits and results of calculation are used for establishment of their distribution near NPP. Water-soluble and water-insoluble radionuclide fractions in the airborne effluents of NPP were investigated. The possibility to evaluate the deposition factor of water-soluble and water-insoluble radionuclides from the NPP jet by comparing the experimental data in the NPP stack and the environment is shown.

Key words atmosphere • environment of Ignalina NPP • numerical models • radionuclide • water-insoluble fractions • water-soluble fractions

Introduction

Physical and chemical properties of radionuclides determine their behavior on their way from a source – the NPP reactor – to biological objects in the environment [2]. Improvement in measurement techniques of radionuclide activity concentrations in the environment and numerous results increased the possibilities to use radionuclides as tracers for horizontal turbulent movement in the atmosphere [3].

Radionuclide concentrations and their forms are measured in the ventilation air in the NPP stack, in the ground level air and deposits. Their transportation in the NPP jet is calculated using semi empirical formulas, which described the turbulent diffusion in the ground level air.

Method

The air sampling equipment for continuous collection of airborne aerosols and deposits is mounted 3.5 km SE from NPP (Fig. 1). The distance between the station of the Institute of Physics (FI station) (E 26° 35' 10.9", N 55° 34' 54.9") and Ignalina NPP (E 26° 33' 29.4", N 55° 36' 24.8") is optimal for the radionuclide registration from NPP in air and deposits. The north-west and north north-east wind direction is prevalent in the region and transport of contamination from NPP to the station is frequently observed. The high airflow rate of 1800 m^3/h through the vinylpolychloride Petrianov filters with the 0.7 square meters area is used for air sampling. The filters were pressed into the pellets of stable geometry and measured by the gamma-spectrometry method.

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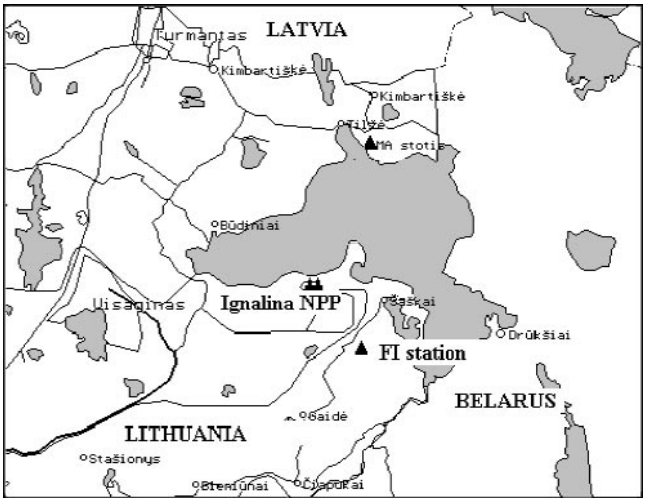

Fig. 1. Location of the geophysical station and the Ignalina NPP.

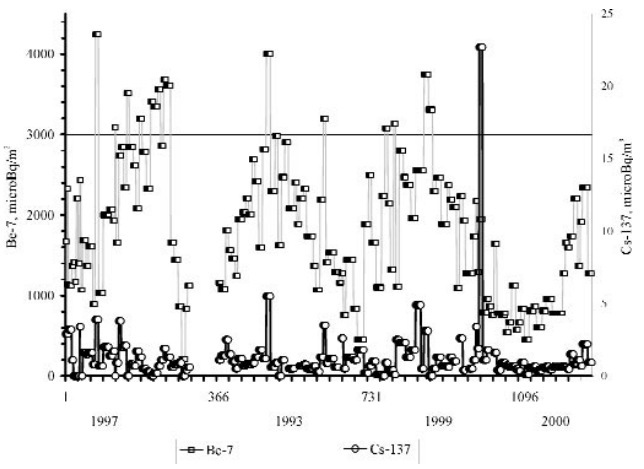

Fig. 2. The ^{137}Cs (primary scale) and ^{7}Be (secondary scale) activity concentrations in air from January 1997 to July 2000.

At the geophysical station, precipitation is gathered from a sloping 10 square meters large surface (snow samples are analysed in winter). A sample for the determination of radionuclide amount in precipitation was prepared nearly every month. Water-soluble and water-insoluble fractions were analysed separately. The method used in our laboratory for concentrating radionuclides (concentrating was performed in 80 L of water applying $Fe(OH)_3$ for setting) allows to collect a number of naturally existing radionuclides in a single sample. This rapid precipitation method was used for the determination of natural radionuclides, activation and fission products. Gamma-radiation of the ashed samples is measured by a high purity Ge well-type detector shielded from the environment.

Radionuclide concentrations and their forms in the air borne effluents of NPP were investigated with a high pressure transportable compressor constructed specially for this purpose that can pump up to 1 m^3 of ventilation air from the NPP stack into steel flasks. During pumping, the air was filtered through aerosol filters FPM-15-1.5 and active carbon filters. Gamma spectra of the flasks and filters were measured using a gamma-spectrometer. The aerosol filters were extracted with deionized water. After 18 hours of extraction, the water-soluble fraction was separated using membrane (0.45 μm) filters.

Results and discussion

In this paper, we discuss the results of systematic measurements of radionuclide activity concentration in air and deposits from January 1997 to July 2000. The ^{137}Cs and ^{7}Be concentrations in air are presented in Fig. 2. The lower detection limit of radionuclide activity concentration in the air was 0.1 μBq/m^3. This limit was caused by the registration criteria of full-energy peaks in energetic gamma-spectra of samples in the areas of radiation of different radionuclides. In the applied software for gamma-spectra expansion, the accepted criteria allowed to estimate only peaks registered with a 10% statistical error. Often to satisfy these conditions the registration time of spectra was extended to 2 days. A full error of establishing air volume, duration of exposition and measuring was smaller than 10%. The error of measurements of radionuclide activity concentrations in the air was 15% for concentrations close to the detection limit and 10% for larger concentrations.

An increase of concentrations of ^{7}Be is observed normally in early summer, and it may be seen in Fig. 2. Descending of stratospheric air to the boundary layer causes separate increases of concentrations. The change of ^{137}Cs concentration depends on a general mixing process of global air mass and Ignalina NPP influence. An increase of ^{137}Cs concentration on September 15–23, 1999, was higher than the change affected by global process. The meteorological data showed that ^{137}Cs could be not from NPP. This increase of ^{137}Cs concentration can be explained by the ^{137}Cs transport from the highly contaminated areas of the Chernobyl region.

The results of radionuclide concentrations in the air samples, in which NPP radionuclides were observed, are presented in Table 1. The radiation of ^{60}Co was registered in 18% of samples, which was related to the frequency of wind direction from NPP. Other radionuclides from NPP were registered episodically.

Solubility in water of the radionuclides emitted from NPP was analyzed in four samples of aerosol air filters. The mean results are presented in Table 2. About 80% of the activation products, ^{60}Co and ^{54}Mn were registered in the water-insoluble form. The radionuclide activity concentration in the deposits is given in Table 3. The lower detection limit of radionuclide activity concentration in the deposits was 0.01 Bq/m^2/month. An increase of ^{7}Be concentration is observed in summer. ^{137}Cs and ^{7}Be concentrations are in good correlation. This showed that the majority of ^{137}Cs in the deposits is of global origin. The ^{60}Co and ^{54}Mn radiation was registered episodically. They were mostly in water-insoluble fraction.

The measurements in the Ignalina NPP stack were performed for the quantitative and qualitative evaluation of the radionuclide source. Results of measurements are presented in Table 4. The lower detection limit of radionuclide activity concentration in the airborne effluents was 0.01 Bq/m^3. Radiation of iodine isotopes during the measurements was not registered. Solubility of radionuclides was analyzed in four aerosols filter samples of the NPP stack.

Table 1. Radionuclide activity concentrations in air.

Date of sampling	Activity concentration, μBq/m^3							
	^{7}Be	^{137}Cs	^{134}Cs	^{54}Mn	^{60}Co	^{59}Fe	^{95}Zr	^{95}Nb
1997.02.10 – 02.18	1690	1.6	<0.1	<0.1	0.7	<0.1	<0.1	<0.1
05.13 – 05.19	2840	2.0	<0.1	1.0	1.6	<0.1	<0.1	<0.1
05.19 – 05.26	2340	2.1	<0.1	1.3	1.5	<0.1	1.7	0.3
06.02 – 06.11	2850	0.8	<0.1	<0.1	0.6	<0.1	<0.1	<0.1
06.11 – 06.18	2610	0.7	<0.1	<0.1	0.6	<0.1	<0.1	<0.1
06.18 – 06.26	2080	1.7	<0.1	0.8	1.3	<0.1	<0.1	<0.1
06.26 – 07.03	3200	1.3	<0.1	<0.1	0.8	<0.1	<0.1	<0.1
07.03 – 07.13	2790	0.5	0.1	0.7	1.0	0.4	<0.1	<0.1
07.13 – 07.23	5330	0.3	<0.1	<0.1	0.3	<0.1	<0.1	<0.1
08.08 – 08.15	3560	0.7	<0.1	0.1	0.6	<0.1	<0.1	<0.1
08.15 – 08.22	2860	1.1	<0.1	0.5	0.6	<0.1	<0.1	<0.1
09.16 – 09.27	1440	0.8	<0.1	<0.1	0.5	<0.1	<0.1	<0.1
1998.02.16 – 02.27	1940	1.2	<0.1	<0.1	0.5	<0.1	<0.1	<0.1
05.16 – 05.23	2980	0.9	<0.1	<0.1	0.3	<0.1	<0.1	<0.1
06.17 – 07.01	2080	0.5	<0.1	0.1	0.3	<0.1	<0.1	<0.1
07.23 – 07.30	2330	0.8	<0.1	0.4	0.5	<0.1	<0.1	<0.1
07.30 – 08.15	1740	0.5	<0.1	2.2	3.0	1.9	0.9	0.5
08.31 – 09.08	2190	1.3	<0.1	0.4	0.6	<0.1	<0.1	<0.1
09.20 – 10.04	1530	1.2	<0.1	0.3	0.4	<0.1	0.5	0.3
1998.12.26 – 01.02	2500	0.7	<0.1	0.06	0.07	<0.1	<0.1	<0.1
1999.01.02 – 02.12	1660	1.0	<0.1	0.07	0.05	<0.1	<0.1	<0.1
01.25 – 02.02	2240	<	<0.1	0.2	0.2	<0.1	<0.1	<0.1
02.09 – 02.15	2140	0.4	<0.1	0.11	0.22	<0.1	<0.1	<0.1
02.23 – 02.28	3130	0.09	<0.1	<	0.08	<0.1	<0.1	<0.1
03.24 – 04.05	2380	1.3	<0.1	<	0.5	<0.1	<0.1	<0.1
06.04 – 06.16	1935	1.1	<0.1	0.4	0.2	<0.1	<0.1	<0.1
09.07 – 09.10	2170	3.4	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
09.10 – 09.15	1288	1.9	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
09.15 – 09.23	1950	22.7	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
09.23 – 10.03	790	1.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
11.06 – 11.17	709	0.7	<0.1	0.3	<0.1	<0.1	<0.1	<0.1
11.17 – 11.24	670	0.9	<0.1	<0.1	0.3	<0.1	<0.1	<0.1
2000.02.22 – 03.01	810	0.59	<0.1	0.23	0.3	<0.1	<0.1	<0.1
03.07 – 03.25	960	0.42	<0.1	<0.14	0.14	<0.1	<0.1	<0.1
04.27 – 05.07	2200	0.82	<0.1	0.64	0.9	<0.1	<0.1	<0.1
05.07 – 05.13	1370	1.1	<0.1	0.97	1.5	<0.1	<0.1	<0.1
05.18 – 06.01	2340	2.2	<0.1	0.59	0.7	<0.1	<0.1	<0.1

The mean values are presented in Table 5. The results show that radionuclides emitted from NPP are in water-soluble fraction. About 20% of ^{137}Cs were found in water insoluble fraction.

The radionuclides registered in the deposits in the Ignalina NPP environment were mostly in insoluble fraction. The supposition can be made that activation products emitted from NPP in soluble fraction partly pass into insoluble fractions in air.

The atmosphere is the most mobile sphere where aerosol composition and properties can change in a short time. Both water-soluble and water-insoluble aerosols of ^{60}Co carriers produced and released from NPP can be formed according to the anthropogenic aerosol in the ground level air, chemical pollution of the environment landscape and season [3]. The most probable mechanism of the transformation of ^{60}Co carriers from water-soluble to water-insoluble form is due to numerous chemical reactions, especially hydrolysis and oxidation on the surface of particles.

The measurement results of radionuclide concentrations in deposits in only one point of the area of Ignalina NPP can be possibly used to estimate the distribution of radionuclides in deposits for the whole region. For this purpose, intensity of ^{60}Co deposits in most points of the area is calculated using measurements results of radionuclide activity concentrations in the airborne effluents of NPP and meteorological data. Radionuclide transportation in the NPP jet was calculated using the Pasquill-Gifford's method [1]. After comparing results of calculations and measurements, the field of radionuclide concentration in deposits was obtained.

The activity concentration of an individual radionuclide in the air, $q^{r}_{(x,y,z,t)}$, when the wind direction is related with the ordinate axis, is calculated by multiplying the meteorologi-

Table 2. Radionuclide activity concentrations in air (aerosol fraction).

Radionuclide	Concentration in water-soluble fraction, μBq/m^3	Concentration in water-insoluble fraction, μBq/m^3	Percentage quantity in water-soluble fraction, %
^{60}Co	0.90±0.09	3.15±0.08	22
^{54}Mn	0.87±0.09	2.95±0.08	23
^{137}Cs	1.45±0.09	3.25±0.08	32

Table 3. Radionuclide activity concentrations in the deposits.

Date of sampling	Activity concentration, Bq/m^2 /month			
	^{7}Be	^{137}Cs	^{60}Co	^{54}Mn
1997.02.14 – 04.08	3.0	0.6	0.2	<0.1
04.08 – 05.27	29.8	2.4	0.1	0.2
05.27 – 07.03	21.3	0.6	<0.1	0.11
07.03 – 07.29	7.3	0.5	<0.1	0.2
07.29 – 10.09	5.8	0.2	<0.1	<0.1
10.09 – 12.01	1.3	0.2	<0.1	<0.1
12.01 – 1998.03.24	0.8	0.1	<0.1	<0.1
1998.03.24 – 04.21	4.6	0.5	<0.1	<0.1
04.21 – 06.01	4.7	0.5	<0.1	0.2
06.01 – 07.01	41.1	2.2	<0.1	<0.1
07.01 – 07.29	4.0	0.1	<0.1	<0.1
07.29 – 08.30	4.9	0.1	0.2	<0.1
08.30 – 10.06	3.3	0.1	<0.1	<0.1
10.06 – 11.11	1.7	<0.1	<0.1	<0.1
11.11 – 12.15	1.6	<0.1	<0.1	<0.1
12.15 – 03.09	1.6	0.1	<0.1	<0.1
1999.03.09 – 04.05	0.6	<0.1	<0.1	<0.1
04.05 – 04.30	1.2	0.12	1.4	<0.1
04.30 – 06.03	1.9	0.2	<0.1	<0.1
06.03 – 07.08	39.2	3.2	0.1	<0.1
07.08 – 07.31	0.8	0.02	<0.1	<0.1
07.31 – 09.08	4.0	0.10	0.002	<0.1
09.08 – 10.28	3.0	0.13	0.002	<0.1

cal dilution factor $G_{(x,y,z,t)}$ and the emission velocity of the radionuclide through the NPP stack, $Q^{r}_{(t)}$:

$$(1)\quad q^{r}_{(x,y,z,t)} = Q^{r}_{(t)} G_{(x,y,z,t)} F^{r}_{R} F^{r}_{W}$$

Coefficients F_R, F_W, estimate a radionuclide removal through the NPP jet due to the radioactive decay – R and wash out with precipitation – W:

$$(2)\quad F^{r}_{R} = \exp - \frac{\lambda \chi}{u} ,$$

$$(3)\quad F^{r}_{W} = \exp - \frac{\Lambda \chi}{u}$$

where λ is the radioactive decay constant, Λ is the constant of washout with precipitation.

For the description of radionuclide flow onto the ground surface and water surface, $A_{s(x,y)}$ the velocity of dry falls out ν_g, could be used

$$(4)\quad A_{s(x,y)} = \nu_g q^{r}_{(x,y,0)}$$

Radionuclides are a passive admixture in respect to the turbulent movement of air. Thus, the same meteorological dilution factor may be used in calculations of the radionuclide distribution as well as for the evaluation of their forms.

Table 4. Radionuclide activity concentrations in the airborne effluents (aerosol fraction).

Time	Radionuclide activity concentration, Bq/m^3				
	^{60}Co	^{54}Mn	^{94}Nb	^{134}Cs	^{137}Cs
1998.12.16 – 9:20	2.60	0.24	0.48	<0.01	1.40
12.17 – 8:50	6.80	0.27	0.07	<0.01	1.10
1999.04.21 – 9:40	5.50	0.72	0.07	0.07	12.1
04.21 – 11:15	1.60	0.12	0.08	<0.01	0.59
04.21 – 13:30	0.25	0.04	<0.01	<0.01	0.39
04.22 – 8:50	0.38	0.04	0.01	<0.01	0.39
04.22 – 14:30	0.76	0.24	<0.01	0.30	0.56
12.01 – 11:40	2.50	0.45	0.44	0.04	3.40
12.01 – 15:10	0.31	0.03	1.30	<0.01	0.11
12.02 – 9:50	0.36	0.08	0.01	<0.01	0.70
12.02 – 11:15	0.03	0.12	<0.01	<0.01	0.26
2000.04.19 – 12:15	0.11	<0.01	<0.01	<0.01	0.17
04.19 – 14:50	0.40	0.03	<0.01	<0.01	0.05
04.20 – 9:20	0.02	0.01	<0.01	<0.01	0.02

Transition from one chemical form to another can be described by a special coefficient by the analogy with radioactive decay description. The washout constant and velocity of deposition for water-soluble and water-insoluble fraction in principle are different and their evaluation is required.

We have the experimental data and the possibility to make calculations for the evaluation of these constants. The duration of radionuclide transformation from water-soluble into water-insoluble fraction is equated to the time during which wind carries them several kilometers. Constant k_{tn} is considerably bigger than constant of a number of radionuclides. The factor of removing of radionuclides from water-soluble fraction in the NPP jet and their accumulation in water-insoluble fraction can be expressed by the equation.

$$(5)\quad F^{rt}_{R} = \exp - \frac{k_{tn}\chi}{u} ,$$

$$(6)\quad F^{rn}_{R} = 1 - \exp - \frac{k_{tn}\chi}{u} .$$

Here, we have a factor of decrease of radionuclides in water-soluble fraction and accumulation of water-insoluble fraction in the NPP jet and the constant k_{tn} of transformation from soluble into insoluble fraction. Values of the constants can be estimated from the percentage ratio of water-soluble and water-insoluble fraction of an individual radionuclide in air, assuming that the average wind velocity is u = 7 m/s being the most favorable to the process. Thus: for ^{60}Co – $k_{tn}=2.5\times10^{-3}$ 1/s, for ^{54}Mn – $k_{tn}=3.0\times10^{3}$ 1/s, for ^{137}Cs – $k_{tn}=2.8\times10^{-3}$ 1/s.

A special computer program is prepared for the calculation of distribution of radionuclide concentrations according to (1) using effluent capacity in airborne effluents, meteoro-

Table 5. Radionuclide activity concentrations in the airborne effluents of NPP (aerosol fraction).

Radionuclide	Concentration in water-soluble fraction, μBq/m^3	Concentration in water-insoluble fraction, μBq/m^3	Percentage quantity in water-soluble fraction, %
^{60}Co	2.07±0.09	0.014±0.01	99.3
^{54}Mn	0.16±0.03	<0.01	100
^{137}Cs	0.97±0.04	0.21±0.01	78
^{94}Nb	0.40±0.04	<0.01	100

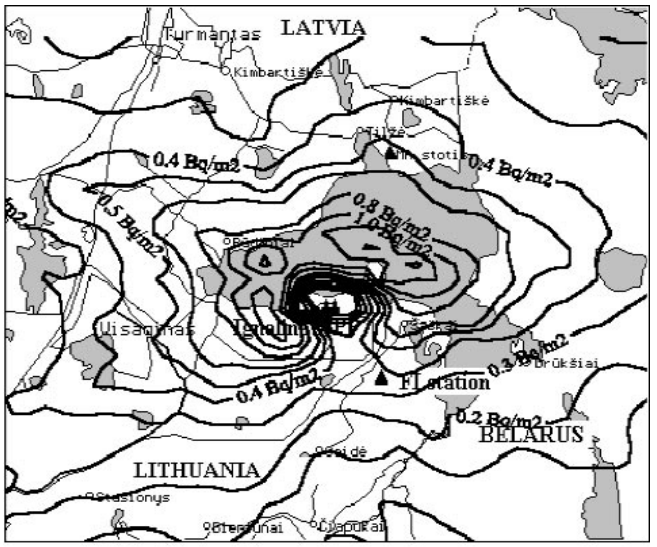

Fig. 3. Distribution of activity concentration of ^{60}Co in deposits in 1997. Calculations were performed using effluent capacity in airborne effluents, meteorological data and experimental results (Table 3).

logical data and the experimental results. The calculation results of distribution of ^{60}Co concentration in deposits are shown in Fig 3. In this calculation, the fraction transformation constant was not evaluated. Meteorological data are constantly measured at the station of the Institute of Physics. Character of distribution of other radionuclides activity concentrations in deposits should be the same. The estimation of the relative quantity of radionuclides in airborne effluents of NPP in 1997 can give distribution of concentration in deposits for these radionuclides.

The experimental data on NPP emitted radionuclides in the air and deposits obtained in 1999 and 2000 (Tables 1, 3) can be used for the estimation of distribution of radionuclides in the deposit fields in the region. It requires processing of a great number of meteorological data.

Conclusions

Results of systematic measurements of radionuclide activity concentration in the air and deposits near the Ignalina NPP from January 1997 to July 2000 are presented. Radionuclides in the airborne effluents of NPP were found in water-soluble fraction. Radionuclides registered in the air and deposits in the Ignalina NPP vicinity were mostly in water-insoluble fraction.

Horizontal turbulent movements in the atmosphere are described using a semi empirical formula of radionuclide transfer in the atmosphere. Radionuclide distribution is calculated applying meteorological information and radionuclide emission data. Data of the Ignalina NPP produced radionuclides; Co^{60} and Mn^{54} in deposits and results of calculation are used for establishment of their distribution in the region.

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