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In situ study of the ^{41}Ar plume released from the Ignalina NPP

Research Article

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Abstract: The ^{41}Ar gamma ray radiation was registered using the in situ method in the vicinity of the Ignalina nuclear power plant (NPP). The sum of gamma rays, that are reaching the HPGe detector, situated along the wind direction, from a number of plume segments and which are registered in the energy range of about 1.29 MeV, is calculated. An independent technological regime of the operating reactor method of the determination of the ^{41}Ar emission rate from NPP stack is introduced.

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1. Introduction

The Ignalina Nuclear Power Plant (NPP) has two RBMK graphite-moderated channel-type 1500 MW reactors: the Unit 1 reactor was shut down in 2005 and the Unit 2 reactor is in operation until 2010. Noble gas radionuclides (Kr, Xe) are produced in nuclear fuel pellets as ^{235}U fission products and ^{41}Ar, the dominating radionuclide in airborne effluents, is formed as the neutron activation product in the reactor active zone and in the air [1].

The ^{41}Ar activity concentrations in the effluents of the NPP during its normal operation may reach several hundreds of Bq m^{-3} [2]. Since the linear attenuation coefficient of the ^{41}Ar ($t_{1/2} = 1.5h$) 1.293 MeV gamma rays radiated in the air is about 0.0069 m^{-1} [3] and 10% of originally radiated gamma rays may reach the 300 m distance from the radionuclide decay point, the ^{41}Ar gamma radiation can be registered *in situ* in the vicinity of the NPP. Analogous measurements of the noble gas radionuclide radiation field and plume geometry were carried out in Belgium and France [4, 5].

The aim of this work is to apply the new calculation method of the radionuclide gamma ray efficiency registration in the NPP plume and to determine the ^{41}Ar emission rate from NPP stack. The local semi-empirical Pasquill-Gifford's model of the radionuclide dispersion in the ground level air was used to determine the plume geometry [6, 7]. The large amount of meteorological data needed for the calculation were provided by the Ignalina NPP meteorological station up to 2005 and later from the HYSPLIT model archive database [8].

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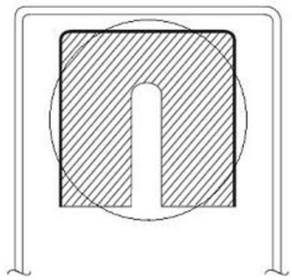

Figure 1. The layout of the ORTEC sensitive volume HPGe detector. *D* is the diameter of the sphere identical to sensitive detector volume, P_d is the mean gamma ray range in the Ge crystal.

2. Method

The gamma ray radiation of ^{41}Ar, released from the Ignalina NPP 150 m height ventilation stack, is registered using the HPGe spectrometer at 1-3 km distance leeward from the source.

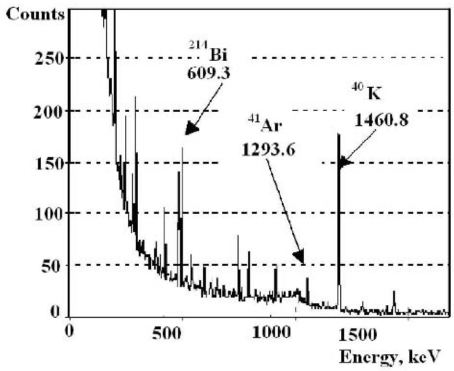

Figure 2. Gamma ray energy spectra obtained during the *in situ* measurement in the vicinity of the Ignalina NPP on July 19, 2006.

The plume geometry is described by the coordinate system which rotates in reference to the wind direction. The x axis is linked with the wind direction, the z axis is linked with the height above the ground level and the y axis is perpendicular to the wind direction. The ground surface at the release point (the middle distance between the Ignalina NPP reactors 1 and 2 ventilation stacks or only stack of reactor 2 after stopping unit 1) corresponds to the origin of coordinates (x_0, y_0, z_0).

The activity concentration of an individual radionuclide in the air, $C_{(x,y,z)}$, at steady meteorological conditions may be calculated using the well-known semi-empirical Pasquill-Gifford's equation:

$$C_{(x,y,z)} = Q\frac{\exp -\frac{\lambda x}{u}}{2\pi\sigma_y\sigma_z u}\exp\left[-\frac{y^2}{2\sigma_y^2}\right] \times \left\{\exp\left[-\frac{(z-h_{ef})^2}{2\sigma_z^2}\right]+\exp\left[-\frac{(z+h_{ef})^2}{2\sigma_z^2}\right]\right\}, \quad (1)$$

where Q is the radionuclide emission rate from the NPP stack, [Bq s^{-1}], u is the wind velocity, [ms^{-1}], λ is the radionuclide decay constant, [s^{-1}], σ_y and σ_z are standard horizontal and vertical deviations of the admixture dispersion in the air, [m], h_{ef} is the effective radionuclide release height, [m].

The gamma radiation from the decay of ^{41}Ar, which is distributed in the plume, is studied using the portable ORTEC HPGe detector placed on the x axis at a measurement point described by a set of coordinates (x_1, 0, 0). ^{41}Ar gamma rays can reach the detector in wide distances from the radionuclide decay points in the plume. The probability of registering gamma rays at different distances to the detector, A_{ef}, can be described as the angle for gamma rays possible to reach detector:

$$A_{ef} = \frac{D}{L_{(x,y,z)}(1-e^{-\eta P_d})}, \quad (2)$$

where D is the diameter of the sphere which is identical to the sensitive detector volume (Fig. 1), [m] (0.040 m), $L_{(x,y,z)} = \sqrt{(x-x_1)^2+(y-y_1)^2+(z-z_1)^2}$ is the distance between the radionuclide decay point and the measurement point (x_1, y_1=0, z_1=0), [m], η is the linear attenuation coefficient of ^{41}Ar radiated gamma rays in the Ge crystal, [m^{-1}] (2.4×10^{-3}), P_d is the mean gamma ray range in the Ge crystal, [m] (0.034 m).

The ^{41}Ar gamma ray radiation can be registered in the ground-level air after the shutdown of the Ignalina NPP Unit 1 reactor (only Unit 2 in operation) (Fig. 2).

An integral over the plume volume, V, from which gamma rays can reach the detector with regard to the radionuclide distribution in the plume, the attenuation in the air and geometrical gamma rays registration efficiency, can be calculated as the sum of gamma rays reaching the detector from the plume with the specified precision of registration in ^{41}Ar photopeak, N_{ef} :

$$N_{ef} = \int_V C(x,y,z)e^{-\tau L(x,y,z)}(A_{ef})dxdydz, \quad (3)$$

where $C_{(x,y,z)}$ is the ^{41}Ar activity concentration in the air, [Bq m^{-3}] at the radionuclide decay point, τ is the linear attenuation of gamma rays coefficient in the air ([m^{-1}]).

The method of the approximate integration is proposed. The triple integral is divided into n equal parts and, thus, it is expressed in sums. The calculation algorithm is based on the square interpolation equation:

$$N_{ef} = Q\sum_{i=1}^{n}\sum_{j=1}^{n}\sum_{k=1}^{n}(C(x_i, y_j, z_k)e^{-\tau L(x_i,y_j,z_k)}[A_{ef}]\Delta_z\Delta_y\Delta_x, \quad (4)$$

where Δx, Δy, Δz are interpolation steps, 10 m. The calculation is performed using the MathCAD software.

The sum of gamma rays reaching the HPGe detector, situated along the wind direction, from a number of plume segments and which are registered in the energy range of about 1.29 MeV, is calculated. This result, obtained using models of the radionuclide spatial distribution in the NPP plume at steady meteorological conditions, provides a tool to establish the relationship between the radionuclide emission from the NPP stack and the appropriate energy gamma ray count rate.

Standard horizontal and vertical deviations of the admixture dispersion in the air are determined using the temperature gradient in the ground-level air, calculated at two measurement heights 2 and 40 m, the average wind velocity and wind direction [6]. These real time meteorological parameters are provided by the meteorological station of the Ignalina NPP and the online meteorology archive data of the HYSPLIT model.

3. Measurement results

Five successful experiments for "in-situ" measurements in Ignalina NPP were carried out. Portable high-resolution gamma-ray spectrometer was used. The location for the measurement was chosen leeward from NPP ventilation stack (Fig. 3). The measuring sites were chosen by taking into mind the current data from the meteorological station of Ignalina Environmental Safety Laboratory.

The wind direction in respect of the measuring site can be presented graphically on the map of the Google Earth software that is interconnected with the HYSPLIT [7] model graphic user interface. Coordinates of the measuring site and distance from emission point can be estimated accurately.

Meteorological parameters (wind direction, velocity, air temperatures at the height of 2 m (T_1) and 40 m (T_2)) for each of experiment were provided by the Ignalina NPP meteorological station until 2005 and later from the HYSPLIT model archive database (Table 1).

The atmosphere stability class obtained using wind velocity and temperature gradient $\Delta T = (T_1 - T_2)/(h_1 - h_2)$. Dispersion for atmosphere stability class D are evaluated

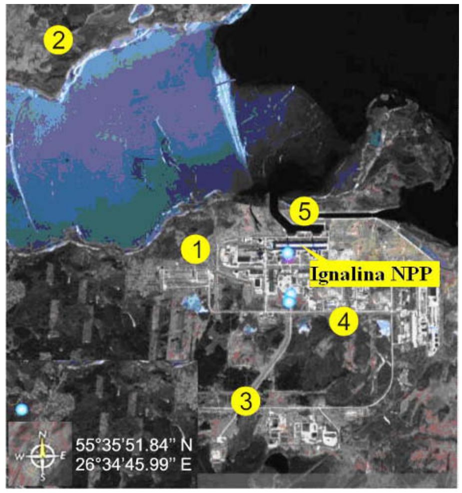

Figure 3. The location of the emission source (Ignalina NPP) and measuring sites.

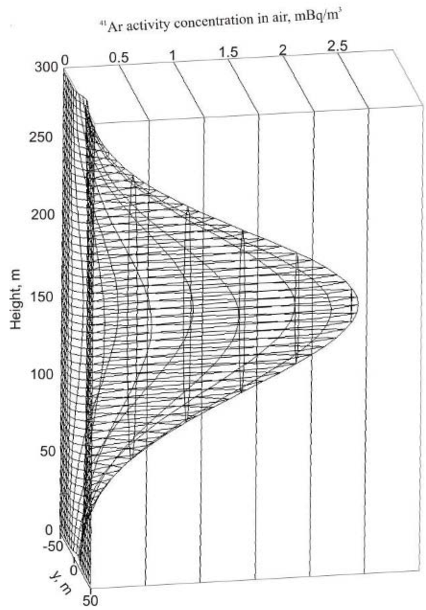

Figure 4. Distribution of ^{41}Ar activity concentration in air, 18-May-00.

using standard dispersion evaluation equations. The meteodata set, given in Table, allows the calculation of the concentration of radionuclides in air, knowing the average radionuclide emission for several hours. Eq. (3) link the registered ^{41}Ar radiated gamma ray count rate with ^{41}Ar emission throughout the stack.

The ^{41}Ar activity concentrations in air and emission rates are calculated using MathCAD software. First 1000 m^3 volume segment V_1 (closest to detector) was approximated

Table 1. Date, geographical location and metrological conditions in measuring site.

	Date	Distance R [m]	Wind direction [deg]	Wind velocity [m/s]	T_1 [°C]	T_2 [°C]	Stability class	σ_z [m]	σ_y [m]
1	18-May-00	1000	106	2.6	19.1	19.6	D	70	50
2	19-May-00	4550	293	6.6	20.5	22.1	D	150	350
3	17-Aug-02	1900	191	3.5	3.0	4.0	D	55	140
4	19-Jul-06	850	134	3.4	20.7	22.4	D	25	65
5	24-Aug-07	450	14	3.4	9.8	10.4	D	15	35

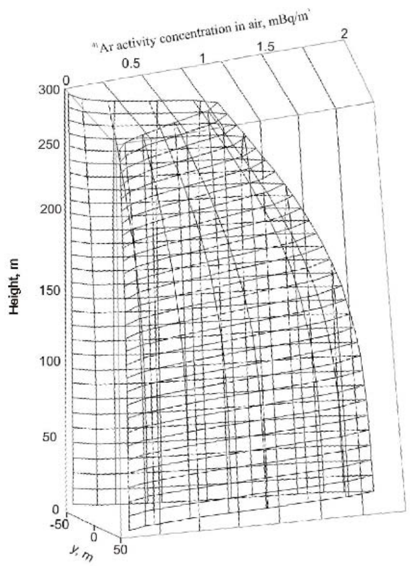

Figure 5. Distribution of ^{41}Ar activity concentration in air, 19-May-00.

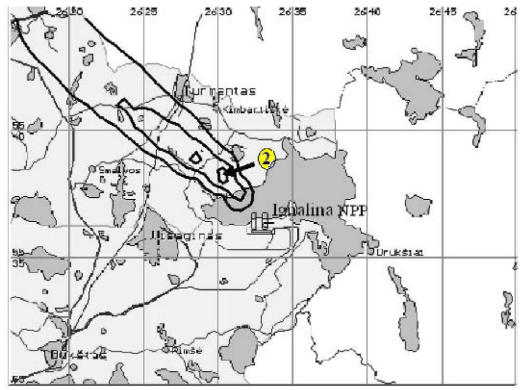

Figure 6. Illustration of 2^{nd} measuring site calculation ^{41}Ar activity concentrations in ground level air.

using a 6.23 m diameter sphere and it was separated to even volume segments of 1 m^3, 10 m^3 and 100 m^3 surrounding detector to specify the registration efficiency of the gamma rays incoming from each of the volume segments (Eq. 1). Total evaluations of gamma rays, incoming from a volume from which gamma rays can possibly reach the detector were estimated with a 10% uncertainty. Calculation step $\Delta = 10$ m in x, y and z directions were used, the ^{41}Ar radiation, which reaches the detector from separate 1000 m^3 volume segments registering efficiency is evaluated. Calculation boundaries in z direction ranged from 5 m to 300 m in x and y directions from -150 m to +150 m. Count ratio of gamma rays P from first 1000 m^3 volume segment and total volume are evaluated. The emission rate is calculated using both experimental and calculated results.

Measurement data of the in situ experiment leeside of the Ignalina NPP and the modeling results are given in Table 2.

Table 2. Results of measurements and calculations.

	Date	Count rate [s^{-1}]	P	C [mBq/m^3]	C_{max} [mBq/m^3]	Emission rate Q [Bq/s]
	1	2	3	4	5	6
1	18-May-00	0.008	0.037	0.06	2.6	1.5E+05
2	19-May-00	0.008	1.43	0.02	1.3	3.0E+06
3	17-Aug-02	0.051	0.19	0.2	45.4	8.1E+06
4	19-Jul-06	0.010	0.00003	1.0	4.0	3.7E+04
5	24-Aug-07	0.010	0.0061	2.3	68.0	7.9E+07

The count rate in the selected region of the gamma ray energy spectrum in the energy range of about 1.29 MeV averaged over 5 experiments carried out in 3 hours of measurements are given in column 2. Calculated ratio of gamma rays P from first 1000 m^3 volume segment and from total volume are presented in column 3. C, [mBq/m^3]

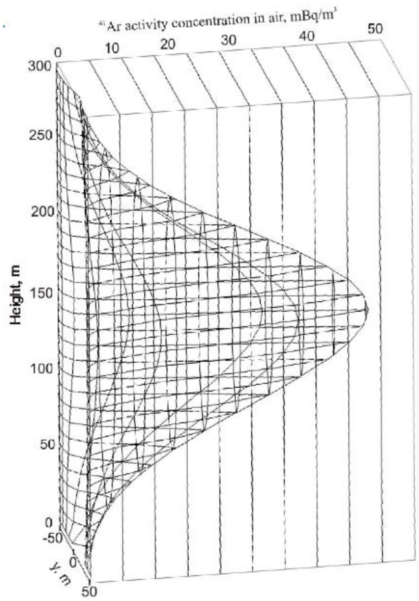

Figure 7. Distribution of ^{41}Ar activity concentration in air, 17-Aug-02.

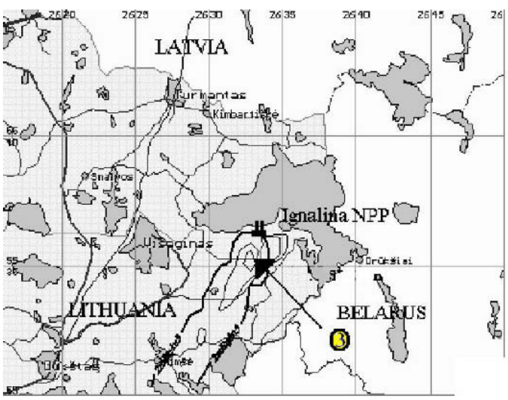

Figure 8. Illustration of 3rd measuring site calculation ^{41}Ar activity concentrations in ground level air.

calculated (Eq. 1) for measurement points described by a set of coordinates (x_1, 0, 0) are presented in column 4. The ^{41}Ar activity concentration C_{max}, mBq/m^3 calculated for the plume axis at a distance from the stack that is the same as the measuring site are presented in column 5. The ^{41}Ar emission rate Q, [Bq/s], is presented in column 6. This value was obtained by selecting it for calculations of the count rate of gamma rays averaged to correspond to measured values presented in column 2.

The results in Table 2 shows, that the set of stabile meteorological parameters described the distribution of ^{41}Ar in the plume corresponding to model (Eq. 1) and quantum

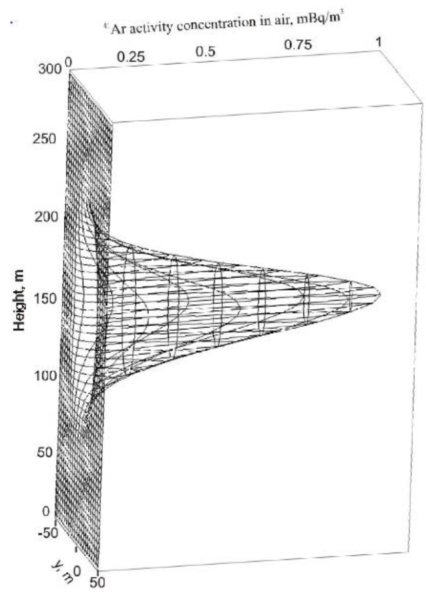

Figure 9. Distribution of ^{41}Ar activity concentration in air, 19-Jul-06.

gamma rays emitted from any point of the plume registration efficiency evaluation. The summarizing over total plume volume enables the direct relationship between the number of counts (column 2) of the appropriate energy gamma rays and the ^{41}Ar emission from the NPP stack (column 6) to be established. This could be evaluated as the basis of the new method for radionuclide emmision from the stack by field measurements.

^{41}Ar distribution in plume, as the middle result of calculation using MathCad software is of particular interest. The variety of ^{41}Ar distribution in the plume is evaluated for measuring periods presented in figures below. Distribution of ^{41}Ar activity concentration in air on plane perpendicular wind direction at measuring sites for all experiments is given in Fig. 4, 5, 7, 9, 10. Distribution of ^{41}Ar activity concentration in ground-level air are for two experiments on 19-May-00 and 17-Aug-02 are calculated (Fig. 6 and 8).

In the second experiment the distance from detector to emission point was large enough for the plume to reach ground level (Fig. 6). Calculated number of gamma rays from first 1000 m^3 volume segment was higher than from total volume. In the third experiment the calculated numbers of gamma rays from first 1000 m^3 volume segment and from total volume (Fig. 8) were comparable. In other cases, only these gamma rays were registered, which came from remote areas which were near the plume axis and the radionuclide activity concentrations were higher.

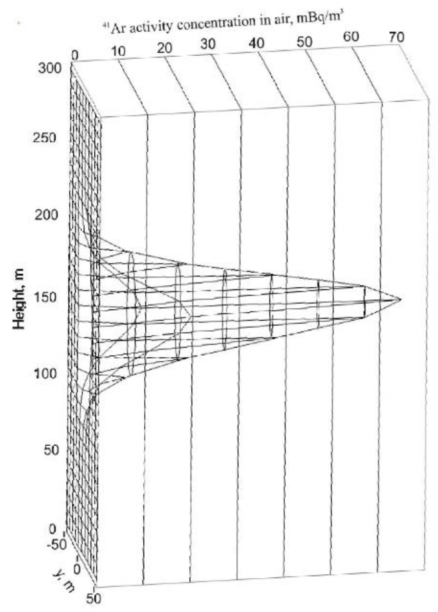

Figure 10. Distribution of ^{41}Ar activity concentration in air, 24-Aug-07.

Results make it possible to estimate radionuclide activity concentration in air and radiation dose of NPP airborne effluents by direct measurement "in-situ" method. The possibility of registering the short-lived radionuclide *in situ* in the optimally chosen measuring site in the environment of the NPP and calculation radionuclide activity concentrations provides the basis to evaluate the air ionization processes as well the formation of N_2O, O_3 and the fine aerosol in the NPP release plume.

4. Conclusions

A new method of calculating registration efficiency for in-situ measurement of gamma ray emitters near the source is given. The set of stabile meteorological parameters described the distribution of ^{41}Ar in the plume and gamma ray quantum emitted from any point of the plume registration efficiency evaluation. Summarizing over total plume volume enables the direct relationship between the number of counts of the appropriate energy gamma rays and the ^{41}Ar emission from the NPP stack to be established. This could be evaluated as the basis of the new method for radionuclide emission from the stack by field measurements.

Five in-situ experiments were conducted registering ^{41}Ar radiation in Ignalina NPP vicinity. It is shown that using high-resolution a portable Ge detector to register radiation of short-living noble gases in NPP environment and real-time meteorological data it is possible to estimate the emission of ^{41}Ar from the source.

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