

The possibility of determining radioactivity of the emissions from atomic electric power plants based on the microwave radiation at 1420 MHz frequency

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The paper describes a passive method of determining the radioactivity level in the emission plumes of atomic power plants (APP) by the secondary emission of radiation from the plume. The emissions have been analyzed from the APPs that exploit the most common types of reactors, and the main beta-radionuclides involved in the emission have been determined. Taking into account the process of electron breeding in the atmosphere, the total energy spectrum of electrons has been obtained. Basic physicochemical reactions were considered resulting in the atomic hydrogen formation in the zone of the emission plume. The spatial distribution of atomic hydrogen concentration in the plume was calculated and the power of emission at 1420 MHz frequency from the volume was estimated. It has been shown that in estimating the radiation impact on the atmosphere we should consider many electron generations because they have mean energy values far exceeding the ionization potentials for atoms and molecules of the air components. It was found that the power emitted by 1 km³ volume at 1420 MHz frequency in the APP emissions can reach $\sim 10^{-13}$ W. The comparison has been carried out with the results of field experiments based on detecting radiation at 1420 MHz from the emission plume of a nuclear reprocessing plant, from whence it follows that the above method enables one to reliably detect the total radioactivity of the APP emissions.

Introduction

In recent years, the problem of radioactive contamination of ground atmospheric layers by emissions of industrial pollutants from the nuclear power plants has become an urgent ecological problem. As a rule, such emissions deliver a number of radioactive elements to the environment, and the total emission radioactivity becomes of the primary significance. The techniques for its determination can be either direct, based on recording the intensities of α , β , and γ radiation, or indirect ones using the effects manifested in the atmosphere under the action of radioactivity. These effects can produce zones of increased ionization, or the zones of increased concentration of gases generated in the atmosphere due to photochemical reactions. These gases can be tracers of the radioactivity and can be recorded either directly or by their characteristic emissions.

The attempts to use the radar techniques for remote monitoring of the zones of enhanced ionization failed because at low contamination levels, when the ion concentration is close to the background and is 10^4 – 10^7 ion/cm³, the sensitivity of standard radars is insufficient. It should be noted that the radar signals create high electromagnetic background that may be of ecological hazard for the territory.

In some papers^{1,2} the authors considered the technique of remote monitoring of radioactivity of

the emissions from radiochemical plants (RCP), based on the atomic hydrogen radiation at 21-cm wavelength, formed in the emission activity and can be recorded remotely in a real time mode. Emissions from nuclear power plants differ greatly from the RCP-emissions both by the isotopic composition and by specific activity (by an order of magnitude).

The goal of this study is to consider the potential of the passive technique of diagnostics of the radioactivity of the APP emission plumes, based on the selection of an atomic hydrogen radiation line in the microwave range. In doing this research job the following problems have been solved: comparison of the emissions from APPs exploiting reactors of different types and a choice of radionuclides making the main contribution to the emission activity; the calculation of energy spectrum of electrons generated in the atmosphere under the action of radiation; modeling of the processes of atomic hydrogen generation in the emission plume and estimation of its radiation intensity.

1. Composition of emissions from the nuclear power plants

The most active sources of radioactive aerosol contamination of the environment from the plants of nuclear-fuel cycle (NFC) are the APPs and RCPs. The other plants of NFC emit to the atmosphere smaller amount of radioactive components.^{3,4} The

radionuclide composition of emissions depends on the type of reactor operated and other its characteristics. Thus, the emissions from APPs and RCPs differ by the isotope composition and also by the presence of liquid and solid components in the RCP emissions. The determined values of emissions for nuclear reactors are given in Table 1.⁴

Table 1. Estimation of annual outcome of emissions into the atmosphere of two reactors with the electric power of 1000 MW

Radionuclide	BWR reactor emissions (reactor with boiling water), Bq/year	PWR reactor emissions (reactor with water under pressure), Bq/year
⁴¹ Ar	$9.25 \cdot 10^{11}$	$9.25 \cdot 10^{11}$
^{83m} Kr	$< 3.7 \cdot 10^{10}$	$3.7 \cdot 10^{10}$
^{85m} Kr	$5.55 \cdot 10^{12}$	$5.2 \cdot 10^{11}$
⁸⁵ Kr	$1.073 \cdot 10^{13}$	$1.739 \cdot 10^{13}$
⁸⁷ Kr	$7.4 \cdot 10^{12}$	$1.11 \cdot 10^{11}$
⁸⁸ Kr	$8.88 \cdot 10^{12}$	$8.51 \cdot 10^{11}$
^{131m} Xe	$6.66 \cdot 10^{11}$	$3.034 \cdot 10^{12}$
^{133m} Xe	$< 3.7 \cdot 10^{10}$	$4.44 \cdot 10^{12}$
¹³³ Xe	$1.184 \cdot 10^{14}$	$4.44 \cdot 10^{14}$
^{135m} Xe	$2.738 \cdot 10^{13}$	$< 3.7 \cdot 10^{10}$
¹³⁵ Xe	$4.07 \cdot 10^{13}$	$3.182 \cdot 10^{12}$
¹³⁸ Xe	$5.18 \cdot 10^{13}$	$< 3.7 \cdot 10^{10}$
¹³¹ I	$1.11 \cdot 10^{10}$	$9.25 \cdot 10^8$
¹³³ I	$9.07 \cdot 10^{10}$	$8.51 \cdot 10^8$
¹⁴ C	$3.515 \cdot 10^{11}$	$2.96 \cdot 10^{11}$
³ H	$1.591 \cdot 10^{12}$	$4.07 \cdot 10^{13}$

As is seen from Table 1, the main amount of the radioactive emissions is due to RNG – radioactive noble gases Ar, Kr, and Xe. These gases and radioactive iodine, whose emissions make up only a small fraction of the total amount of the emissions, are the main sources of radioactive impact on the atmospheric components. At a continuous operation the nuclear power plant is a stationary source of gas–aerosol emissions to the atmospheric boundary layer. Thus, the radioactive gases and iodine isotopes play a leading role in the radiation situation in the areas around nuclear power plants. For this reason the calculations were made just for such a set of β -active radionuclides.

2. Overall energy distribution of electrons in the emission plume

Because of the beta decay of the radionuclides fast (high-energy) electrons are generated, which ionize the medium, and cause the molecular dissociation. In this paper the process of energy degradation of fast electrons in the air is considered within the framework of the following model: the basic mechanism of electron generation is ionization; electron–electron and electron–ion impacts can be neglected; the dissipative processes are not taken into account; atoms of media before the collisions are at rest.

The succession of determining the overall electron energy spectrum is as follows:

1) the spectrum of electrons generated in the air is being determined (spectrum of electrons of beta-decay for each radionuclide is known⁵);

2) the energy spectrum is determined of the primary electrons thermalizing in the air;

3) the spectra of secondary electrons are found; further the procedure by points 2, 3 is repeated for the next generations of electrons;

4) the overall energy spectrum of electrons is found.

The calculation of energy distribution of electrons in the homogeneous medium with the uniformly distributed sources was made using the initial equation describing the energy degradation⁶:

$$\Sigma(E)\Phi(E) - \int_E^{E_0} \Sigma_s(E' \rightarrow E)\Phi(E')dE' = S(E), \quad (1)$$

where Σ , Σ_s are the integral and differential macroscopic section of energy transfer⁴ obtained by multiplying the appropriate macroscopic sections by the concentration of atoms in a homogeneous medium:

$$\Sigma_s(E' \rightarrow E) = \frac{d\sigma}{de} N,$$

where $\varepsilon = E' - E$ is the energy lost by a particle at a single interaction; $S(E)$ is the electron spectrum of beta decay, electron/cm³·s; $\Phi(E)$ is the density of the electron flux with the energy E , electron/cm²·s.

The calculated results from Ref. 3, show that the mean energy of the primary and secondary electrons essentially exceeds (by about 10⁵ times) the ionization potentials of atoms and molecules of the air, which are ~15 eV. The increase of the number of electrons of different generations will continue until the electrons are able to sustain the ionization process. Hence, it is necessary to take into account the contribution of different electron generations. Taking into consideration the ionization potential of the air, the calculation was made up to the ninth electron generation.

Because of electron identity, the overall electron energy distribution can be obtained by summing the energy spectra of electrons of different radionuclides and generations. Figure 1a shows the overall energy distribution of the electron number density for isotopes under consideration.

Figure 1 shows that the electron quantity with relatively low energies < 100 keV represents a considerable fraction from the total electron quantity. As an illustration, Fig. 1b shows the distribution of the energy spectra of electrons with low energies. These electrons make a considerable contribution in the amount of bremsstrahlung – basic mechanism of radiation impact on air components. Under its impact the photochemical reactions occur resulting in the atomic hydrogen formation, which

emits spontaneously at 21 cm wavelength (1420 MHz).

A sharp decrease, observed at low energies, is due to the fact that in this case it is necessary to consider the effects of electron adhesion, which are not taken into account within the framework of an accepted model.

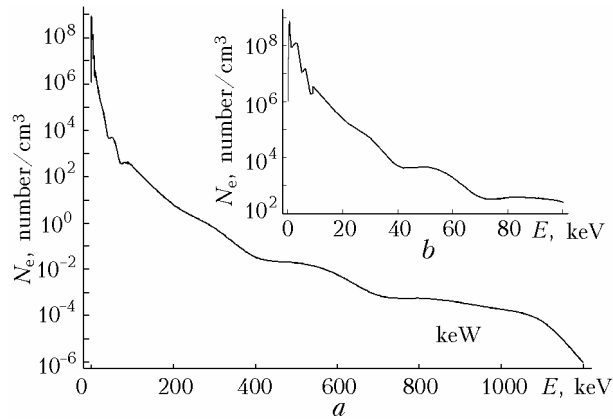


Fig. 1. Overall energy spectrum of electrons.

3. Photochemical reactions resulting in the formation of atomic hydrogen

Below 10-km height the atomic hydrogen concentration in the atmosphere is negligible, because the radiation responsible for photolysis of atmospheric gases with the formation of $H(\lambda < 2900 \text{ \AA})$ is fully absorbed by the ozone layer located at 10–30 km altitudes. According to literature,⁷ the formation of atomic hydrogen in the standard lower atmosphere is due to the photolysis of the following compounds: methane CH_4 , ammonia NH_3 , molecular hydrogen H_2 , formaldehyde $HCHO$, and water H_2O . For the reactions of the atomic hydrogen formation to occur in the atmosphere the radiation is necessary with the wavelengths λ from 844.7 to 3500 \AA . The dissociation energy of hydrogenous air components is calculated by the formula $E_d = hc/\lambda$. This amount of energy is within the limits from $2.352 \cdot 10^{-11}$ – $5.676 \cdot 10^{-12}$ erg.

We consider that the basic impact of radionuclides on the air components is the bremsstrahlung of β -electrons. It initiates a series of photochemical reactions including the formation of atomic hydrogen. Below we calculate the bremsstrahlung intensity taking into account the overall energy electron spectrum.

4. Assessment of plume radiation intensity

To calculate the energy emitted by an electron in a given spectral range, we use the conclusions of classical electrodynamics.⁸ In this case, assuming that the spectrum of electron bremsstrahlung is characterized by the homogeneous energy distribution, the electron–atom collisions are rare (as

compared with the frequency of emission of electromagnetic waves), the scattering event itself is very fast as compared with the period of the field oscillation, and we can write the formula for the energy amount emitted by an electron in a frequency interval dv per 1 s, when retarded, in the following form:

$$dQ_v = \frac{8e^2v^2}{3c^3} v_{\text{eff}} dv,$$

where e is the electron charge, c is the speed of light, v is the electron velocity, $v_{\text{eff}} = Nv\sigma_t$ is the effective collision frequency (N is the number of atoms in 1 cm^3 , σ_t is the transport cross section of energy transfer).

The scattering is considered to be isotropic, the stopping time of one electron is 10^{-6} s and taking into account the results presented in Fig. 1, after summing up we obtain that the energy emitted in the spectral range from 844.7 to 3500 \AA will be 0.131 erg/cm^3 or $8.2 \cdot 10^{10} \text{ eV/cm}^3$.

Based on the above assessments, we can say that the decay of the above-mentioned isotopes in the zone of emission plume yields about 10^{16} hydrogen atoms per m^3 per second. The equilibrium concentration of H atoms in this case will be determined by the processes of creation and recombination. If we know the recombination rate, we can assess the atomic stationary concentration from the following equation⁷:

$$N_H = \sqrt{\frac{N_{H(\text{unsteady})}}{2k}}, \quad (2)$$

where N_H is the stationary concentration of hydrogen atoms, k is the recombination coefficient 10^{-13} – $10^{-14} \text{ m}^3/\text{s}$. Equation (2) is valid, when the recombination is a basic mechanism of disappearance of hydrogen atoms, that is typical for atmospheric conditions. In this case the stationary H concentration in the plume is about 10^{15} m^3 .

To evaluate the spatial distribution of radioactivity in the APP emission plume the Puckvill–Gifford calculation model was used. This model is widely used to calculate the isotope concentration in the atmosphere at distances from 10 to 12 km with the wind direction from the source. According to Ref. 9 the general formula for calculation of the mean particle concentration in the emission plume is of the form:

$$q_A(x, y, z) = \frac{Q}{2\pi\sigma_y(x)\sigma_z(x)U_{sr}} \exp\left[-\frac{y^2}{2\sigma_y^2(x)}\right] \times \left[\exp\left[-\frac{(z-h_{\text{eff}})^2}{2\sigma_z^2(x)}\right] + \exp\left[-\frac{(z+h_{\text{eff}})^2}{2\sigma_z^2(x)}\right] \right] \text{Bq/cm}^3, \quad (3)$$

where Q is the emission source power, Bq/s; x is the coordinate measured from the source along the direction of mean wind, m; y is the coordinate

perpendicular to the direction x , m ; z is the vertical coordinate, m ; U_{sr} is the wind velocity averaged over the mixing layer, m/s ; h_{eff} is the effective emission height, m ; σ_y , σ_z are the variances (standard deviations) along the corresponding directions.

To determine the supplementary altitude of jet rise due to thermal and dynamic factors the admissible relation is presented in Ref. 9:

$$\Delta h = A_1 \left(2.61 \frac{\sqrt{Q_h}}{u_h} + 0.029 \frac{u_s D_0}{u_h} \right),$$

where Q_h is the thermal source power, kW ; u_h is the wind velocity at the emission height, m/s ; u_s is the rate of gas flow from the source outlet, m/s ; D_0 is the diameter of the source outlet, m ; A_1 is the dimensionless parameter determined by the class of atmospheric stability (see Table 2).

Table 2

Class of stability	A	B	C	D	E	F
A_1	2.6	2.1	1.6	1.1	0.9	0.7

For a homogeneous flat terrain of the locality, the data are sufficient on the wind velocity at two levels: at the height of wind vane u_1 and at the height of a possible air jet rise or emission u_h . The vertical wind profile is expressed as $u_z = u_1(z/z_1)^m$. The value of the parameter m for the height of the layer 10–500 m at $z_1 = 10$ m (the wind vane height) depending on the class of stability are in Table 3.

Table 3

Class of stability	A – B	C	D	E	F
m	0.1–0.12	0.13	0.13–0.15	0.16–0.50	0.7–0.8

The calculation was carried out for the initial data given in Table 4. Because the atomic hydrogen concentration is directly proportional to the emission activity, i.e., the amount of radioisotopes emitted to the atmosphere, the formula (3) can be used for calculating the distribution of the stationary concentration H in the air with the account of Eq. (2).

Table 4. Initial data used in the calculations

Atmospheric stratification	Neutral
Ventilation tube: height diameter	100 m 1 m
Mean wind velocity in a direction of impurity transfer (at 1-m height, at wind vane height of 10 m, at emission height)	5 m/s
The rate of air (gas) outflow from the tube outlet	5 m/s
Type of radioactive impurity	elementary impurity (atomic)

Figure 2 shows the distribution of atomic hydrogen stationary concentration in the emission plume of the atomic power plant at different heights.

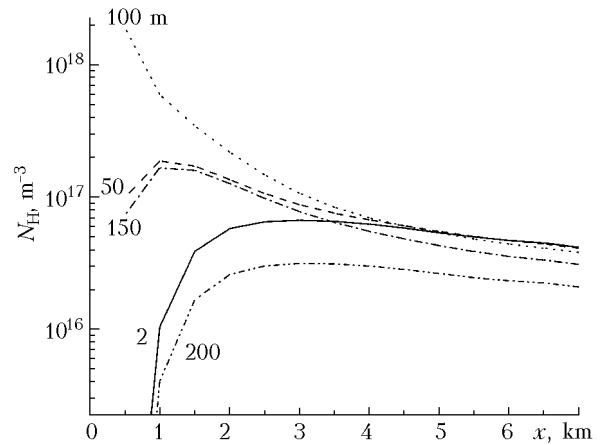


Fig. 2. Distribution of atomic hydrogen stationary concentration in the atmosphere in the case of emissions from APP with the reactor of BWR type.

Figure 2 shows that the maximum atomic hydrogen concentration can be obtained at distances from 0.5 to 3 km at the parameters specified. At distances exceeding 3 km the atomic hydrogen concentration at the above-mentioned heights becomes approximately the same and equals $5 \cdot 10^{14} m^3$. The air volume, containing atomic hydrogen, can reach $\sim 10 km^3$.

Now we determine the radiation intensity at 1420 MHz frequency from the zone of emission plume. The radiation intensity at single transition is determined by the probability of the radiation transition A_{nk} and can be determined by the classical formula: $S = E_{nk} A_{nk}$, where $E_{nk} = h\Delta\nu$ (h is the Planck's constant, $\Delta\nu$ is the transition frequency). According to literature data¹⁰ the energy of hyperfine splitting of the ground energy level of atomic hydrogen is $\sim 9.412 \cdot 10^{-23} J$ for $\Delta\nu = 1420.4037517 MHz$. The probability of the transition A_{nk} is equal to $3 \cdot 10^{-15} s^{-1}$. Then the power of emission by an atom due to single transition S equals $2.824 \cdot 10^{-37} W$.

Because the atomic hydrogen concentration in one cubic meter of emission plume is 10^{15} atoms, then the power, emitted by the volume of 1 km^3 at 1420 MHz frequency, equals $1.5 \cdot 10^{-13} - 9.592 \cdot 10^{-14} W$. The radiation at the above frequency was detected in the field experiments for the case of the nuclear reprocessing plant² from the distance $\sim 25 km$. The limiting sensitivity of a radiometer used in the experiments was $10^{-17} W$. Thus the calculated value of the radiation intensity for the nuclear power plant (NPP) emissions and a comparison of these emissions with the results for the nuclear reprocessing plant^{1,2} show that it is possible to use the radiation at 1420 MHz frequency for detecting the radioactivity level of NPP emissions using the existing radiometers.

Conclusion

The calculations performed demonstrate that for evaluating the radiation impact on the atmosphere it is necessary to consider many electron generations because they have mean energy values far exceeding the ionization potentials of atoms and molecules of the air components.

In spreading of the plume in the atmosphere, large volumes of atmospheric air undergo active irradiation, and the emitting volume may reach 10 km^3 and more. The region of maximum atomic hydrogen concentrations in the emission plume can be determined by modeling the impurity transport.

The power emitted by a 1 km^3 volume of the APP emissions at 1420 MHz frequency may reach $\sim 10^{-13} \text{ W}$. The existing radiometers have made it possible to detect the general level of radioactivity of APP emissions at the distances 20 to 25 km.

References

1. S.T. Penin and L.K. Chistyakova, Atmos. Oceanic Opt. **10**, No. 1, 45–49 (1997).
2. V.Yu. Chistyakov, D.V. Losev, L.K. Chistyakova, S.T. Penin, Yu.K. Tarabrin, V.P. Yakubov, and L.A. Yuriev, J. Microwave and Opt. Technol. Lett. **16**, 225–260 (1997).
3. G.A. Kolotkov, S.T. Penin and L.K. Chistyakova, Proc. SPIE **6160**, Chapter 2, 84–96 (2005).
4. V.M. Kolobashkin, P.M. Rubtsov, P.M. Ruzhanskii, et al., *Radiation Characteristics of Irradiated Nuclear Fuel* (Energoatomizdat, Moscow, 1982), 146 pp.
5. S.V. Avakyan, P.N. Il'in, V.M. Lavrov, and G.N. Ogurtsov, *Cross Sections of Processes of Ionization and Excitation of UV Radiation at Electron Ion Photon Collisions with Atoms and Molecules of Atmospheric Gases* (GOI, St. Petersburg, 2000), 365 pp.
6. A.M. Kolchuzhkin and V.V. Uchaikin, *Introduction into the Theory of Particle Penetration Through Matter* (Atomizdat, Moscow, 1978), 255 pp.
7. Kh. Okabe, *Photochemistry of Small Molecules* (Mir, Moscow, 1981), 504 pp.
8. Ya.B. Zeldovich and Yu.P. Raizer, *Physics of Shock Waves and High-Temperature Hydrodynamics Phenomena* (Phys.-Math. Literature Press, Moscow, 1966), 702 pp.
9. K.P. Makhon'ko, ed., *Manual on the Organization of Control of State of Natural Medium in the Area of APP Location* (Gidrometeoizdat, Leningrad, 1990), 266 pp.
10. I.S. Shklovskii, *Cosmic Radiation* (Gostekhizdat, Moscow, 1956), 341 pp.