FORMATION AND DYNAMICS OF RADIATION EMITTED BY ATOMIC HYDROGEN IN THE EMISSION PLUMES OF NUCLEAR PLANTS

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*We propose a technique for ecological monitoring of the atmospheric emissions from nuclear plants by detecting emission of the atomic hydrogen at 1420 MHz frequency (*21.1 cm *wavelength). Atomic hydrogen is the product of interaction between radioactive elements and atmospheric gases and particularly , with the water vapor. We estimate the amplitude of the signal due to atomic hydrogen radiation as well as the influence of the atmosphere on the emission plume dispersal and on the volume concentration of hydrogen atoms. The relationships have been derived allowing determination of optimum geometric conditions for detecting radiation under different meteorological conditions.*

INTRODUCTION

The emission of atomic hydrogen occurs due to hyperfine splitting of the ground energy level of H atom into two sublevels. The cause of splitting is interaction of spins of atomic nuclei and electron. The energy level at parallel orientation of electron and nuclear spins is somewhat higher than that at antiparallel spin orientation. At a spontaneous change of the electron spin orientation to the opposite one, a quantum of the frequency 1420.40575 MHz is emitted. It is assumed, that the atomic hydrogen concentration in the emission plume should be increased due to a radioactive irradiation of large volumes of the atmospheric air.[1]

In this paper, we analyze photochemical processes resulting in the atomic hydrogen generation in a radioactive emission plume from a nuclear-processing plant and the background radiation power in the atmosphere as well as the influence of atmospheric conditions on the spread of the radiating plume. The background radiation power has been estimated which includes both the galactic hydrogen radiation and that from hydrogen, generated in the atmosphere due to photochemical processes. Processes are considered that cause generation, the lifetime and dynamics of the atomic hydrogen concentration. Based on this analysis we examine a possibility of detecting a signal against the background noise.

SOURCES OF GENERATION AND BACKGROUND CONCENTRATION OF ATOMIC HYDROGEN IN THE ATMOSPHERE

Atomic hydrogen as well as the hydroxyl OH refer to the family of odd hydrogen. High reactivity of these components makes them very essential partners of chemical processes in the atmosphere. The lifetime of hydrogen radicals depends considerably on the altitude (Fig. 1).[2]

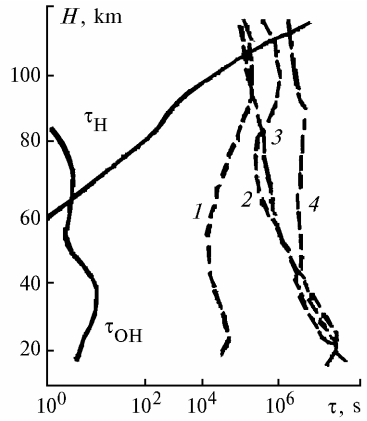

FIG. 1. Photochemical lifetimes of H (t_H) *and* OH (t_{OH}) *and time constants of zonal (1), meridional (2), and vertical (3) transfer and one-dimensional diffusion (4).*

The lifetime of all the components of the family is less than the transfer time constants, therefore the concentration of hydrogen radicals is determined by photochemical and not by dynamic processes.

In addition, the content of these components strongly depends on the solar radiation flux, (day time and season, respectively) since the basic source of their generation is photodissociation of the long-lived hydrogen components. Figure 2 shows vertical distribution of the concentration of hydrogen radicals, calculated for midday in March.[2]

In the upper atmosphere (thermosphere) atomic hydrogen is a dominating component of the entire family, whereas, below the level of 75 km its concentration is much less than the concentration of other hydrogen components.

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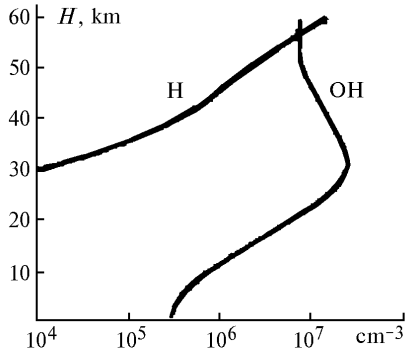

FIG. 2. The model distribution of concentration (cm^{-3}) *of atomic hydrogen* H *and radical* OH.

Below the level of 10 km the concentration of atomic hydrogen in the atmosphere becomes negligible since the radiation responsible for the photolysis of atmospheric gases yielding hydrogen atoms ($\lambda < 2900$ Å) is completely absorbed by the ozone layer located at the altitudes[3] from 10 to 30 km.

Consider now the sources of the atomic hydrogen in the standard lower atmosphere. Based on numerous measurements of chemical composition of the atmosphere,[4] we can conclude that the generation of atomic hydrogen in the 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. The total concentration of molecules of these components in the atmosphere is 10^{19}–10^{20} $1/m^3$. In Ref. 5 the data on photochemistry of such compound obtained by different authors are generalized. We only consider the reactions yielding the atomic hydrogen and consider the conditions favoring them.

The primary processes of photolysis of the methane under the action of radiation at $\lambda = 1100$–1600 Å can be represented as the following reactions:

$$CH_4 \xrightarrow{h\nu} CH_3 + H$$
$$CH_4 \xrightarrow{h\nu} CH_2 + H_2 + H_2 \xrightarrow{h\nu} H + H$$
$$CH_4 \xrightarrow{h\nu} CH + H_2 + H,$$

in this case, the absorption spectrum in this range is continuous.

For ammonia the three primary processes take place at radiation photolysis in the near and vacuum ultraviolet.

$$NH_3 \xrightarrow{h\nu} NH_2 + H \qquad \lambda < 2800 \text{ Å} \qquad (1)$$
$$NH_3 \xrightarrow{h\nu} NH + H \qquad \lambda < 2240 \text{ Å} \qquad (2)$$
$$NH_3 \xrightarrow{h\nu} NH + H + H \qquad \lambda < 14570 \text{ Å} \qquad (3)$$

Processes (1) and (3) are most probable when the dissociation is excited by light at $\lambda < 2800$ Å.

The primary photochemical process causing the photodissociation of formaldehyde is the reaction:

$HCHO \xrightarrow{h\nu} H + HCO$ (the threshold wavelength $\lambda = 3500$ Å).

For H_2O molecules the primary process causing the photodissociation in the wavelength range from 1050 to 1900 Å is the generation of H and OH:

$$H_2O \xrightarrow{h\nu} H + OH \qquad \lambda < 2420 \text{ Å}.$$

Molecular hydrogen generated due to secondary reactions, as compared with the above ones, can dissociate along with the molecules of atmospheric hydrogen under the action of radiation in the wavelength range from 844.7 to 1108 Å due to the reaction

$$H_2 \xrightarrow{h\nu} H + H.$$

Thus, we can conclude that the radiation at $\lambda = 844.7$–3500 Å makes the largest contribution to the process of atomic hydrogen generation. The equilibrium concentration of the H atoms will be determined in this case by the processes of creation and recombination.

FORMATION AND DYNAMICS OF RADIATION EMITTED BY ATOMIC HYDROGEN IN THE EMISSION PLUME

The characteristic property of emissions from processing of nuclear fuel is emission of radioactive isotopes of a series of elements in the emission plume. Analysis of nuclear plants operating in a closed cycle has shown that the most significant from the radioactive elements emitted is the isotope ^{85}Kr. The annual emission of ^{85}Kr is about $1.6 \cdot 10^7$ Curie for radiochemical plants with the productivity of 1500 tons per year.[6] For a source with standard parameters (the stack altitude $H = 200$ m, the diameter $D = 2$ m, the speed of emission $V = 10$ m/s) the above productivity results in the radioactivity of every m^3 of the emission is $6 \cdot 10^8$ Bq. Since ^{85}Kr is a β – active isotope, the result of its decay is the emission of about ($6 \cdot 10^8$) electrons per second with the energy of 0.67 MeV. In the process of the emission dispersal its expansion takes place because of vertical and horizontal diffusion and wind. The results of calculation of the average distribution of concentration of ^{85}Kr depending on the distance to the source are presented below.

To estimate the concentration of radionuclides in the plume we used a standard model of Gaussian diffusion from a continuous source.[2] This model has been selected since it is widely used in modern practice for development of the governmental standards setting the limits on atmospheric pollution by industrial emissions. According to this model the impurity concentration N in the emission plume is expressed by the formula:

$$N = \frac{Q}{2\pi\sigma_y \sigma_z u}\left[\exp\left(-\frac{(z-H)^2}{2s_z^2}\right) + \exp\left(-\frac{(z+H)^2}{2s_z^2}\right)\right] \times$$

$$\times \exp\left(-\frac{y^2}{2s_y^2}\right), \quad (4)$$

where Q is the source power; $\sigma_y = \sigma_y(x)$, $\sigma_z = \sigma_z(x)$ are horizontal and vertical variances of impurity concentration.

The values of σ_y and σ_z for $10^2 < x < 10^4$ m are calculated by Briggs formula[2]: $\sigma_y(x) = \sigma_\theta x/(1+c_2x)^{1/2}$, $\sigma_z = a_1 x^{b_1}/(1 + a_2 x^{b_2})$, $\sigma_\theta = c_1(\tau/\tau_0)^{0.2}$, ($\tau$ is the averaging time, $\tau_0 = 10$ min); H is the source height; u is the wind velocity.

The values of a_1, a_2, b_1, b_2, c_1 and c_2 are shown in Table I.

TABLE I.

Atmospheric stratification index	σ_y		σ_z			
	c_1	c_2	a_1	a_2	b_1	b_2
3	0.11	10^{-4}	0.1120	$9.05\cdot10^{-4}$	0.920	0.718
4	0.08	10^{-4}	0.0980	$1.35\cdot10^{-3}$	0.889	0.668
5	0.06	10^{-4}	0.0609	$1.96\cdot10^{-3}$	0.895	0.684

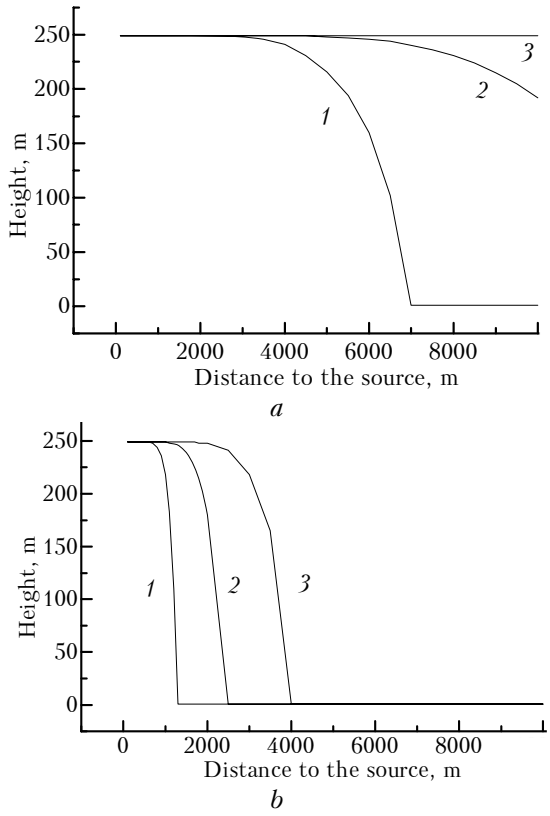

FIG. 3. Dependence of the maximum concentration level height of radionuclides in a plume on the distance to a source for various atmospheric stratification indices: n = 3 (1); n = 4 (2); n = 5 (3); a is the smooth underlying surface; b is the uneven surface: wood or suburb.

Figure 3 shows the dependence of the altitude of the maximum impurity concentration on the distance to the source for a vertical cross-section of the plume at different stratification of the atmospheric boundary layer. It follows from these data that the plume portion from 500 m to – 10 km from the source is most favorable for observations.

It should be noted that the surface roughnesses due to the presence of trees and buildings makes a significant impact on the plume emission propagation. Such roughness can be taken into account by changing the parameter of surface roughness, whose magnitude affects the values of σ_z, σ_y in the expression for the Gaussian diffusion model (Eq. (4)).

Figure 4 shows the plume shape at the roughness parameter characteristic of the underlying surface covered with wood and for the suburb. In this case, the value of σ_z is calculated by the expression

$\sigma_z = a_1 x(1 + a_2 x)^{b_2}$.

The parameters in the expression for σ_z, σ_y are taken from Table II.

TABLE II.

Atmospheric stratification index	σ_y		σ_z		
	c_1	c_2	a_1	a_2	b_2
3	0.22	$4\cdot10^{-4}$	0.20	0	0
4	0.16	$4\cdot10^{-4}$	0.14	$3\cdot10^{-4}$	-0.5
5	0.11	$4\cdot10^{-4}$	0.08	$1.5\cdot10^{-4}$	-0.5

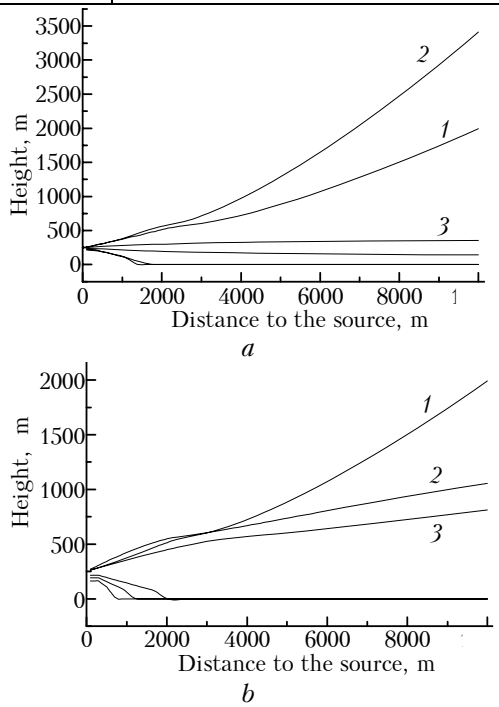

FIG. 4. Contours of the emission plume at a level 1/e of the maximum concentration of radionuclides for various parameters of stratification of the atmosphere: n = 3 (1); n = 4 (2); n = 5 (3); a is the smooth underlying surface; b is the uneven surface: wood or suburb.

All the calculations have been carried out for a moderately unstable, neutral and moderately stable stratifications (the Pasquill-Terner stability classes, $n = 3, 4, 5$, respectively). As follows from the data of meteorological observations for the state of the atmospheric boundary layer,[2] the operation conditions at $n = 3$ most frequently occur in the unstable climatic periods, spring and fall.

The decrease of the size of optimal plume region is obvious as compared with its dimensions characteristic of uniform underlying surface. Due to the plume spread in the atmosphere, a large body of air is exposed to radioactive radiation.

Based on the results obtained with the account for receiving antenna parameters we can determine most suitable location and direction of the antenna for detecting the plume microwave emission under specific meteorological conditions. For a narrow directed antenna with the directional pattern ~ 10° the optimal distance for location is approximately 25–27 km from the emission plume. Such distance allows one to observe the most favorable portion of the plume from 500 m to 10 km, where the atomic hydrogen radiation intensity is maximum.

ESTIMATION OF THE INTENSITY, LIFETIME AND BROADENING OF THE ATOMIC HYDROGEN EMISSION LINE IN THE EMISSION PLUME.

For estimation of the intensity of radiation from a plume, its spectrum and time variation the physical mechanisms of interaction of radionuclei with the atmosphere are considered. As a basic factor of irradiation to the atmosphere we consider the bremsstrahlung of electrons generated in the decay process of the ^{85}Kr isotope. It is known that the bremsstrahlung spectrum is characterized by a homogeneous distribution over energy. The characteristic length where an electron loses its energy[7] is $3\cdot10^4$ cm. To calculate the energy emitted by an electron within a given spectral range, we need to solve a complex quantum – mechanical problem, taking into account all the possible sinks of electron energy due to elastic and inelastic collisions, averaging over all possible excited atomic and molecular states. Such a solution would be too cumbersome and unsuitable for numerical estimates. In this case, the result suitable for estimates can be obtained, based on the classical electrodynamics. Since the scattering process too fast as compared to the period of field oscillations, one can write the formula for the amount of energy emitted by an electron when decelerating to the frequency range from ω to $\omega + \Delta\omega$ in the form (see Ref. 7)

$$d\varepsilon_\omega = \frac{2}{3\pi}\frac{e^2}{c^3}(\Delta\mathbf{v})^2\, d\omega, \qquad (5)$$

where e is the electron charge, c is the light speed; $\Delta\mathbf{v}$ is the variation of the velocity vector in scattering. This formula is valid in the frequency range from ω_{max}, which in this case is 10^{21} l/s. In order to find the energy Q_ω, which an electron emits into the spectral range $d\omega$ per 1 s, it is necessary to sum $d\omega$ over all collisions.

Taking the value of the collision frequency to be of the order of gas kinetic[7] one (10^{14} s^{-1}) and considering the scattering to be isotropic, after summation we have $Q_\omega = 1.5\cdot10^{-2}$ erg/s. This means that during the time of deceleration of one electron (-10^{-6} s) the energy $1.5\cdot10^{-8}$ erg or 10^4 eV will be emitted into the spectral range from $\lambda = 844.7$ Å to 3500 Å. It should be noted that in this case the absorption coefficient of the compounds considered for a given spectral range is on the average 500 atm$^{-1}\cdot$cm^{-1}. For a comparison, the absorption coefficient of molecular nitrogen N_2 in some narrow bands may amount to the values of 50 to 100 atm$^{-1}\cdot$cm^{-1}. Based on the estimates presented it is hoped that as a result of the decay of the ^{85}Kr isotope in the region of emission plume the H atoms can be generated at a rate of $1.5\cdot10^{12}$ m$^{-3}\cdot$s^{-1}.

The recombination rate for H atoms and OH radicals can be estimated from the equation[5]:

$$\frac{dc}{dt} = 2\,k\,c^2, \qquad (6)$$

where c is the atom and radical concentration, 1/m^3, k is the recombination, m^3/s.

This equation is true for the case when the recombination is the basic mechanism of decomposition. Let us estimate stationary concentration of H in the plume for such a situation. It is known from the experimental data[5] that the recombination rate for atoms of neutral hydrogen under normal conditions is about 10^{-13}–10^{-14} m$^3\cdot$mol$^{-1}\cdot$s^{-1}.

The generation rate of H per m^3 is $I_H \sim 1.5\cdot10^{12}$ (1/m$^3\cdot$s) then, based on the solution of Eq. (6), we obtain the relationship for the stationary H concentration:

$$c \approx \sqrt{I_H/2k} \sim (2.5 - 8.5)\cdot10^{12}\ \text{m}^{-3}.$$

Thus, the concentration of hydrogen atoms in 1 km^3 of the emission plume is about 10^{21}–10^{22} atoms.

Intensity of radiation (line) is determined by the probability of radiation transition A_{nk} and can be calculated by the formula[8]

$$S = 2\pi\,\hbar\,\omega_{nk}\,A_{nk}, \qquad (7)$$

where $2\pi\hbar\,\omega_{nk} = E_n - E_k$ is the quantum energy, $\hbar$ is the Plank constant.

According to Ref. 8 the energy of hyperfine splitting of stable levels of hydrogen atoms is equal to 10^{-24} J for $\Delta\nu\,(F, F^1) = 1420.4057517$ MHz. The transition probability A_{nk} equals $3\cdot10^{-15}$ s^{-1} (see Ref. 10). Then the radiation intensity for one act of emission transmission is $S = 3\cdot10^{-15}\cdot10^{-24} = 3\cdot10^{-39}$ W.

From the above reasoning it is not difficult to calculate that the energy emitted by atomic hydrogen

at the frequency 1420 MHz will be about $2 \cdot 10^{-17}$—$8 \cdot 10^{-18}$W. Such intensity is quite sufficient for detecting the radioactive contamination of the environment at large distances.

Lines of hydrogen atom will be broadened due to the Doppler effect, caused by the progressive motion of atoms. The collisional broadening mechanism is neglected because radiation is due to hyperfine splitting of the energy level. The speed of progressive motion of atoms is determined by the distribution of excess energy at photolysis of hydrogen containing molecules by photofragments. Based on the experimental data[5] of measurements of progressive speed of photofragments, the energy of progressive motion of hydrogen atoms does not exceed 2 eV, hence, the Doppler width of hydrogen line 1420 MHz cannot be larger than 150 kHz.

BACKGROUND RADIATION AT FREQUENCIES 1–3 GHZ

The principal sources of noise making difficult the detection of the atomic hydrogen radiation in the emission plume of a radiochemical plant are the cosmic radio–frequency radiation and the background radiation of the atmosphere in a given spectral range. In radio astronomy, the background radiation power is expressed in terms of its radiation temperature corresponding to the temperature of ideal black body which total radiation power coincides with the noise power. Figure 5 gives the dependence of radiation temperature of the background on the frequency.[9] It is seen that the background is minimal in the spectral range from 1.2 to 10 GHz (λ = 3 to 30 cm). The total power of background at the surface level strongly depends on time, however, the above power does not exceed[2] 10^{-21} W/m^2.

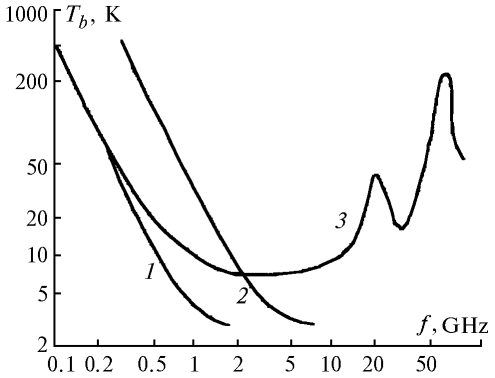

FIG. 5. Dependence of the background radiation on the frequency: cosmic background without the atmosphere (1) and that with the atmosphere (2), and atmospheric background (3).

The radiation attenuation at the frequency of 1.4 GHz due to the radiation absorption in the atmosphere does not exceed 2 dB at different elevation angles of the antenna. In this case, at the elevation angle of 90° the radiation attenuation is 0.03 dB in the calm atmosphere.[10] The presence of background aerosol does not practically affect the signal absorption and under common conditions this background aerosol may be neglected. The values of the linear extinction coefficient in the rain of different intensity from 1.0 to 100 mm/hr according to the data of International Consultative Radio Committee (ICRC) range from 0.002 to 0.01 dB/km. The noise level can considerably be decreased if the measurements are made at night–time when the background due to the radiation of atmospheric hydrogen as well as the solar background are minimal because of high rate of the H atoms recombination.

CONCLUSION

Thus, calculations have shown, that the concentration of atomic hydrogen in the emission plume should be increased due to radioactive irradiation of the atmospheric air and can be up to 10^{21}–10^{22} km^{-3}. The power emitted by atomic hydrogen from one cubic kilometer per one second at the frequency 1420 MHz, should be $2 \cdot 10^{-17} - 8 \cdot 10^{-18}$ W.

The plume when moving in the atmosphere expands due to vertical and horizontal diffusion and under the action of wind. As a result, large volumes of the atmospheric air occupied by the plume and close regions will be exposed to the radioactive irradiation. As a result, the radiating volume can be more than 10 km^3.

Since power of the background radiation at a given frequency with the account for the atmospheric absorption is 10^{-22} W, the use of high-sensitive receivers would allow the detection of radionuclides in the industrial emissions.

Thus, the passive remote sensing technique proposed for detection of activity of the nuclear plants by detecting radiation of atomic hydrogen in the atmospheric emission plume is shown to be promising for practical applications.

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