### Techniques for remotely detecting radioactive anomalies in the near-ground atmosphere

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Active methods of remote detection of radioactive anomalies in the atmosphere, including methods employing the effects arising from the interactions of laser radiation with the products of radiolytic decomposition and at photochemical reactions among atmospheric constituents, are analyzed. The results achieved in developing passive remote technologies for monitoring radioactive pollution due to industrial wastes releases into the atmosphere are considered briefly along with the method of laserinduced breakdown of aerosols. The technologies under consideration are shown to be promising in ecological monitoring of radioactive pollution of the atmosphere. High sensitivity of the methods allows pollutants to be detected from long distances. Simultaneous use of the passive and active methods provides for acquiring more information on the radioactive anomalies examined.

One of the most pressing ecological problems in recent years is the necessity of monitoring radioactive pollution of the near-ground layer of the atmosphere. Remote methods are most promising for this purpose. These methods can be divided into two groups: direct methods based on recording the intensity and spectrum of radiation passed through the atmosphere and indirect methods that employ the effects arising in the atmosphere as it interacts with radiation.

Although the direct methods are more widely used nowadays, their low resolution and sensitivity restrict the distance, from which measurements are possible, to hundreds meters for  $\gamma$ -radiation. As to  $\alpha$ - and  $\beta$ radiations, they cannot be recorded remotely because of their low penetrating power.

Among the indirect methods, the radar method is most highly developed. It is used to measure the level of atmospheric ionization by radiation, and the concentration of charged particles in this method is judged from the reflection coefficient of the sensing microwave radiation (this coefficient is a function of the degree of ionization). 1-4 First data on the radar echoes from radioactive clouds were obtained in 1986 at the Chernobyl nuclear disaster. Then the cycle of experiments in different regions<sup>5</sup> (a total of several hundreds observations) has demonstrated the efficiency of this method in the cases of huge emergency releases of the radioactive decay products into the atmosphere under various weather conditions. The observations were conducted with the use of radars of standard radio altimeters and guidance stations of centimeter and decimeter wavelengths regions. 6

At a low pollution level (the concentration of ions close to the background value and equal to  $10^4$ – $10^7$  ions/cm<sup>3</sup>), the sensitivity of standard radars is insufficient. The ways to improve the sensitivity are now being sought. A possible way is to use microwave compression for obtaining high-power short microwave pulses<sup>4</sup> in order to select signals and decrease the radar "blind zone." However, low resolution of radar methods restricts their capabilities in local radiation monitoring. Also, it should be noted that at systematic measurements the radar signals induce a high electromagnetic background, which itself is an ecological hazard for the territory.

At the same time, other manifestations of radioactivity in the atmosphere are considered from the viewpoint of their use for identification of the radioactivity itself. Among those there are the effects arising from the interactions of laser radiation with the products of radiolytic decomposition<sup>7,8</sup> photochemical reactions of atmospheric constituents.<sup>7,12</sup> Interaction of laser radiation with a radioactive zone includes both the incoherent effects connected with aerosol, molecular, and Raman scattering and the coherent effects of resonance absorption and induced fluorescence. It is worth noting that sensitivity of the techniques that use resonance effects is much higher 7,9 because of larger absorption cross sections. In Refs. 10 and 11 one can find ideas on using microwavecontrolled Brillouin scattering of light from an atmospheric volume ionized due to radioactivity. It was shown that sensitivity of this technique could reach 10<sup>7</sup> ions/cm<sup>3</sup>. However, to use resonance effects, one needs for the wavelength of a laser sources to fall in resonance with the electronic transition of specific gaseous constituents. Besides, these laser sources must have power high enough for operation from long distances and high degree of coherence. The set of these requirements, if to be met simultaneously, makes it difficult to realize these methods.

In this connection, it seems to be timely developing novel techniques of monitoring radioactivity in the atmosphere that would allow measurements in

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real time. The research works in this area are being done at the Institute of Atmospheric Optics of SB RAS in cooperation with the research groups from Tomsk State University, Physical-Technical Institute, and Tomsk Polytechnic University. In this paper I present brief analysis of the results my colleagues and I have achieved in the field of the development of new techniques for studying optical anomalies in the nearground atmosphere due to industrial releases (based on the data of Refs. 12–19).

### 1. Models of atmospheric anomalies arising under the action of radioactivity

It is known that radiation from radioactive elements can cause excitation and ionization of atmospheric constituents. In this case, the environment is affected most strongly by secondary electrons generated at ionization. These electrons, even having the energy less than the ionization threshold of molecules, are capable of exciting the molecules and causing their dissociation.

It is a specific feature of the near-ground atmosphere nearby radiochemical or mining enterprises that ionization due to radioactive releases yields high concentration of molecules of electronegative oxygen and aerosols. The low-temperature weakly ionized medium formed initially in the track region is electrically neutral plasma consisting of both positive and negative molecular and atomic ions. However, due to the processes of electron attachment and adsorption of molecular ions on particles of atmospheric aerosol, a large number of aerosol ions and ion clusters are generated, as a result, the ion-electron plasma transforms into the aerosol one. Inside such formations, charge separation and formation of local geoelectric fields can occur under the effect of the Earth's electric and magnetic fields, as well as gravitation. The possibility of forming the mirror distortions of the ionosphere should be noted as well.

Analysis of the models of radiation effects on the atmosphere showed  $^{3,4,20,21}$  that the overwhelming majority of these models consider a radioactive release as a hard ionizer.

The set of kinetic equations for ions was solved by different authors for different initial conditions, different atmospheric layers, and different sets of plasmochemical reactions (from ten to hundreds) with the made allowance for the track structure of the ionization process, as well as with or without regard for complex and aquated ions. For example, the kinetics of clusters and aerosol formations generated in the lower atmosphere due to radioactive releases was considered in Ref. 20. In this case, diffusion of all particles, drift of charged particles in the local electric field, fall of large particles (or drops), and upward transport of small particles by upwelling air flows were considered.

It was assumed that the recombination rate of small charged particles is constant similarly to the case with elementary ions, and the attachment coefficient depends linearly on the field. It was shown that formation of local anomalous electric fields (up to several  $kV\cdot m^{-1}$ ) is possible in the lower troposphere under the effect of radioactivity. In this case, the charge separation is caused by downward transport of the negative charge by falling drops and by upward transport of the positive charge by upwelling convective flows.

Some investigations showed that strong electric fields in the troposphere affect the state of the ionosphere, giving rise to short-term (less than 4 h) oscillations of the electron density, ion distribution, and temperature. One of the possible mechanisms of this effect is electrodynamics. According to this mechanism, abrupt changes of the vertical electric field in the lower troposphere induce the horizontal field in the ionosphere.

The problem of penetration of the electric field from a source located in the near-ground atmosphere into the ionosphere was considered for the first time in Ref. 23. It was shown that the electric field penetrates into the ionosphere, where its value is  $\sim 1 \text{ mV} \cdot \text{m}^{-1}$ . For the atmospheric electricity this value is negligibly small. However, in the ionosphere it cannot be neglected. In Ref. 22 it was shown that the extent of the distorted region can achieve more than 400 km. The maximum change of the plasma concentration relative to the unperturbed value is roughly 20%, and the horizontal distribution of concentration is characterized by two pronounced focuses (positive and negative distortion).

The distribution of the critical frequency as a deviation from the mean value was measured within several hours after the accident at Three Mile Island nuclear power plant in Pennsylvania in 1979 by the Intercosmos–19 satellite. These measurements were in a good agreement with model calculations. Thus, the models of electric anomalies in the atmosphere under the effect of radioactivity can be used for monitoring of radioactive releases.

In our opinion, the methods of passive detection of neutral and excited atoms and molecules generated in the atmosphere under the effect of radioactivity are promising for detection of radioactive anomalies. 12,13,24 The main requirements to such neutral-particle indicators are stable concentration in the ordinary atmosphere and concentration sensitivity to the effect of radiation.

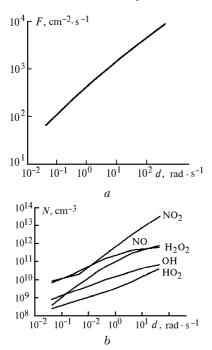
With such an approach, the relaxation processes in the weakly ionized plasma should be considered not only as the processes responsible for ion loss, but also as the processes of generation of neutral components such as the atoms N, O, H, and molecules NO,  $NO_2$ ,  $NO_3$ , radicals OH, etc.

Figure 1 shows the concentration of some neutral components N and the flow density F of radio photons emitted at a transition on the hyperfine structure of the ground state of the OH radical ( $\lambda \approx 18$  cm) as functions of the absorbed dose  $^{25}$ 

$$F(\text{cm}^{-2} \cdot \text{s}^{-1}) = \frac{1}{6} A N_{\text{OH}} R,$$
 (1)

where  $A = 7.7 \cdot 10^{-11} \text{ s}^{-1}$  is the probability of radiative decay of the level, R is the radius of the emitting region.

As seen from Fig. 1 the calculated power density of the radiant flux increases up to  $10^{-25} \ \mathrm{W} \cdot \mathrm{cm}^{-2}$  under condition that a half of the OH radicals is in the radiative state. In addition, the concentration of NO, NO<sub>2</sub>, H<sub>2</sub>O<sub>2</sub>, HO<sub>2</sub> molecules, radical OH, and, as a consequence, of the atomic hydrogen, H, are sensitive to the radiation dose, what makes it possible to use them as indicators of radioactive pollution.



**Fig. 1.** Flow density of radio photons F(a) and concentration of some neutral components N (b) as a function of the radiation dose. Initial conditions: T = 293 K, humidity 90%, exposure time e = 60 s, initial concentration  $N_{\text{(OH)}0} =$  $= 10^6 \text{ cm}^{-3}$ 

Having available reliable data on the radioactivity of releases one can calibrate the equipment for further quantitative measurements of radioactivity in pollution plumes of nuclear cycle plants operating under standard conditions and in case of an emergency.

#### 2. Passive diagnostics of radioactivity in the atmosphere using microwave radiation emitted by atomic hydrogen

Let us consider some results obtained on the concentration of atomic hydrogen and OH radical.

Analysis of literature has shown that the major part of the release from closed-cycle nuclear plants is in  $^{85}\mathrm{Kr}$  isotope. For a plant with the output of 1500 t/yr, the total annual release of this isotope is  $1.6 \cdot 10^7$  Ci (Ref. 26). Assuming that it is emitted from a 200-m high stack 2 m in diameter with the outflow speed of 10 m/s, the radioactivity of every cubic meter of the release is about  $6 \cdot 10^8$  Bq. In this case, decaying  $^{85}$ Kr produces  $6 \cdot 10^8$  electrons with the energy of 670 keV per second in every cubic meter. The generation of electrons causes dissociation of water vapor and produces hydrogen atoms at the concentration of 10<sup>12</sup> m<sup>-3</sup>/s under standard conditions. This yields about  $2 \cdot 10^{-17}$  $8 \cdot 10^{-17} \text{ W} \cdot \text{m}^{-3}$  power of microwave radiation emitted by atomic hydrogen. With time, the plume expands because of vertical and horizontal diffusion and under the effect of wind. As a result, large volumes of atmospheric air occupied by the plume and adjacent to it are subject to radioactive irradiation. Thus, for the emitting volume of 10 km<sup>3</sup> (height of 200-500 m), the total radiation power can be estimated as 10<sup>-8</sup> W. The band width of the signal with the allowance for the Doppler broadening should not exceed 150 kHz. The emission is spontaneous and, therefore, incoherent and nonpolarized. At a distance of 30 km from the plume, this source creates the radiant flux density of  $10^{-18} \ \mathrm{W/m^2}$ , which is detectable by radio astronomic methods. To detect such a source, a receiving system with the effective antenna area of 1 m<sup>2</sup> should have the sensitivity no less than 0.4 K, in terms of the radiative temperature.

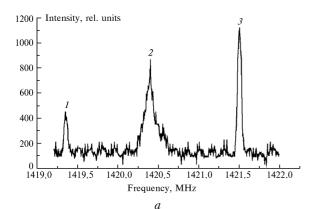
Analysis of the background radio emission of the atmosphere in the studied region has shown that the total power of the noise background at the ground level depends strongly on the time of a day, but does not exceed  $10^{-21}$  W/m<sup>2</sup> (Ref. 27). Atmospheric extinction (due to absorption) of the radiation with the frequency from 1.2 to 1.4 GHz did not exceed 2 dB, and the presence of a background aerosol (for example, rain) has practically no effect on the signal absorption and can be neglected under standard conditions. <sup>28</sup> The noise level is much lower at night, when the noise caused by emission of atomic hydrogen, as well as solar noise, is minimum because of the high recombination rate of hydrogen atoms.

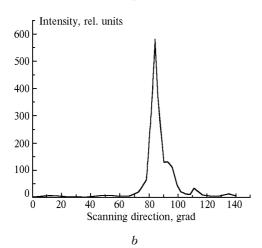
Thus, it is confirmed that emission of atomic hydrogen at the wavelength of 21 cm due to hyperfine splitting of the ground level can be used for detection of radioactivity in pollution plumes of nuclear fuel processing plants, because the concentration of atomic hydrogen in the region of the plume of a normally operating plant (i.e., at the constant absorbed dose) reaches the steady-state level ( $\sim 10^8 \text{ cm}^{-3}$  $d \approx 0.43 \text{ rad/s}$ ) (Ref. 12). As to the OH radical, its concentration along with the flow density of radio photons with the wavelength  $\lambda = 18 \text{ cm}$  strongly depends on the absorbed dose. At the same time, the concentration of OH in the not irradiated atmosphere does not exceed  $10^6$  cm<sup>-3</sup>. Thus, all the requirements to neutral-particle indicators of radioactivity in the atmosphere are fulfilled.

Since H and the OH radical can be considered as a lightweight admixtures in the atmosphere, they does not deposit, but leave the irradiated region only due to diffusion. Consequently, the pollution plume and the adjacent region contain significant amount of OH radicals, whose concentration depends on the plume radioactivity. Since OH and atomic hydrogen emit in the same wavelength region, the instrumentation used for detection of radiation emitted by atomic hydrogen can also be used for detecting the radiation at 18 cm wavelength without any modifications.

The concentration and dynamics of radionuclids in the zone of radioactive deposits and in the plumes of nuclear fuel processing plants were estimated based on the model of Gauss diffusion from a continuous source.<sup>29</sup> For experimental justification of the possibility of recording microwave radiation from a radioactive plume, a special-purpose radiometer was developed. It has the following characteristics <sup>13</sup>:

- sensitivity T = 0.05-0.005 K with the averaging time of 1 s;
- the amplification factor of a low-noise amplifier 60 dB:
  - noise temperature 70 K;
- output temperature drift less than  $0.4~\mathrm{mV/^\circ C}$  in the temperature region from 18 to  $40^\circ\mathrm{C}$ .





**Fig. 2.** Example of the spectrum recorded in the direction to the maximum radiation and coincident with the direction to the radiochemical plant  $^{13}$ : labels at the frequencies of 1419.348 and 1421.505 MHz (1, 3) and signal from the plume (2) (a); radiation intensity distribution in the horizontal direction from 0 (the direction certainly aside the plume) to  $180^{\circ}$  (the direction from the plume at  $\sim 83^{\circ}$ ) (b).

A parabolic antenna was set at an altitude of 20~m above the ground and at a distance of 25--30~km from the source of radioactive pollution. Received signals were recorded and processed on an IBM computer connected to the spectrum analyzer.

For absolute reference of the level of received signals, calibration was performed against the "cold" sky sector. Every measurement session included recording of a signal at azimuth sighting of the antenna in the horizontal plane near the maximum of radiation and calibration measurements with sighting at the cold section with the antenna looking at the azimuth of  $90^{\circ}$  and at an elevation angle of  $60^{\circ}$ . The direction to the cold sector has been taken as a zero azimuth angle. The radiation emitted by atomic hydrogen in the plume was stably observed in the experiment in both the frequency and azimuth.

The measured spectrum of radio-wave radiation at the frequency of 1420 MHz from a nuclear fuel processing plant at the distance up to 25 km and the intensity distribution along the azimuth are exemplified in Fig. 2.

These results indicate that the considered method can be used for ecological monitoring of radioactive pollution of the atmosphere by nuclear plants. High sensitivity of this method allows detection of pollution at long distances from the object. Therefore, the receiving instrumentation can be installed aboard a car, an aircraft, or a satellite.

# 3. Remote detection of uranium pollution of the atmosphere by the methods of active laser sensing (LIBS method)

We have studied the method based on direct analysis of emission spectra of laser-induced plasma and known as LIBS (Laser Induced Breakdown Spectrometry) as a candidate for making remote chemical analysis of radioactive and toxic elements in the pollution plumes of nuclear plants. One of the important advantages of this method is the fact that if the emission spectra of laser plasma are used as indicators, then one performs element analysis, which is independent of structure the element is contained in, because spectral lines of elementary ions are separated. That means that, it does not matter which substance, aerosols or molecular gas species contain the substance sought. Besides, there is no need in special preparation of samples, and remote analysis becomes possible, because the currently available lasers are capable of creating breakdown in air at a distance of 500 m and longer.

Analysis of the data on the composition of gas and aerosol releases from radiochemical plants $^{24}$  has allowed separation of the lines, which can be used for recording, identification, and quantitative chemical analysis, in emission spectra of radioactive elements. The data are given below in the Table.

Table. Fromising times of Tadioactive elements								
	U		Th		Sr		Pu	
	Lines, $\Delta$	U <sub>e</sub> , eV	Lines, Δ	$U_{\rm e}$ , eV	Lines, Δ	U <sub>e</sub> , eV	Lines, $\Delta$	$U_{\rm e}$ , eV
	II 2865.68	4.32	II 2832.32	4.89	II 3380.71	6.61	2939.12	_
	II 3670.07	3.49	II 2837.30	5.14	II 3464.46	6.62	2954.54	_
	II 3854.66	3.79	II 3402.70	4.12	II 4077.71	3.04	I 3851.12	3.22
	II 3859.58	3.24	II 3609.44	3.94	II 4215.52	2.94	I 3895.87	3.12
	II 3890.36	3.22	II 4019.14	3.08	I 4607.331	2.69	II 4472.70	3.02
	II 4090.13	3.24	II 4116.71	3.76	-	_	II 4504.92	3.00
	II 4116.10	3.01	II 4381.66	3.66	-	-	_	-
	Nb		V		W		Mo	
	Lines, $\Delta$	$U_{\rm e}$ , eV	Lines, Δ	$U_{\rm e}$ , eV	Lines, $\Delta$	$U_{\rm e}$ , eV	Lines, $\Delta$	$U_{\rm e}$ , eV
	II 2927.81	4.75	II 2924.03	4.63	I 2944.39	4.57	II 2638.76	6.23
	II 2941.54	4.65	II 2924.64	4.61	I 2946.98	4.57	II 2816.15	6.06
	II 3094.18	4.52	II 3102.30	4.36	I 4008.75	3.45	II 2848.23	5.95
	II 3130.79	4.40	II 3110.71	4.33	I 4074.36	3.40	II 2871.51	5.86
	II 3163.40	4.30	I 3183.41	3.91	I 4294.61	3.25	I 3170.35	3.91
	I 3349.07	3.97	I 3183.98	3.90	_	_	I 3798.25	3.26
	I 3358.42	4.04	I 3185.40	3.96	_	_	I 3902.96	3.17
	I 3580.28	3.59	I 4379.24	3.13	_	_	-	_
	I 4058.93	3.18	-	_	_	_	_	_
	I 4163.47	3.24	_	-	_	_	_	_
	I 4163.66	2.99	-	_	-	-	-	_
	I 4164 66	2.02						

Table. Promising lines of radioactive elements

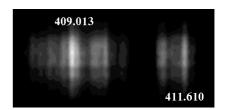
Here  $U_{\rm e}$  is the excitation energy.

Analysis of the technological process enrichment and processing of irradiated fuel<sup>30,31</sup> showed that one of the possible component of a release is UF<sub>6</sub>. It is well-known that UF<sub>6</sub> hydrolyzes by the atmospheric moisture because of its high chemical activity. One of the components produced in hydrolysis is solid-phase aerosol, uranilfluoride UO<sub>2</sub>F<sub>2</sub>. Since UO<sub>2</sub>F<sub>2</sub> well attracts water molecules, its particles in the pollution plume are either in the form of a hydrated solid-phase aerosol or in the form of two-layer particles produced in the humid atmosphere at the growth of droplets on uranilfluoride particles as condensation nuclei. In this case, partial solution of the solid nucleus is possible.

These data have served as a prerequisite for selection of the element U in experimental studies and evaluation of the sensitivity of the considered approach for identification of this element from the emission spectra of aerosol particles containing  $UO_2F_2$ compounds under the effect of pulsed CO2 laser radiation.

The uranium emission was recorded using the spectral line of the uranium ion at the wavelength  $\lambda = 409.013$  nm. This line was selected based on analysis of uranium spectra and the spectra of ordinary atmospheric aerosol containing Ca, Si, and Al elements, which can hamper in situ identification of the uranium lines.

Figure 3a shows the emission spectrum of the breakdown plasma initiated on the UO<sub>2</sub>F<sub>2</sub> particle. One can clearly see the uranium spectral lines, especially the lines brightest in the considered region:  $\lambda = 409.013$  and 411.610 nm.



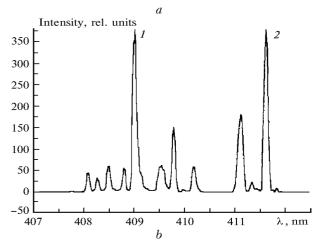


Fig. 3. Image of spectrum displayed on the monitor at TV recording of the emission spectrum of plasma initiated at uranilfluoride particles at the delay  $t_{\rm d}$  = 4.2  $\mu s$ . For the brightest uranium lines, the wavelength is indicated, in nm (a). The spectrum obtained from processing of the recorded image: spectral lines of uranium ions with  $\lambda = 409.013$  (1) and 411.610 nm (2) (b).

The resulting spectrum is shown in Fig. 3b. It is worth noting that almost no background is seen in the figure. This allows even relatively weak uranium spectral lines to be separated out.

This effect is possible because the uranium ionization potential is only 6.2 eV and the laser pulse is rather long (pulse duration  $\sim 1.5~\mu s$ ).

Remote analysis can be performed at the distance up to 1 km from the source of radioactive pollution. In this case, the atmospheric extinction of the signal is insignificant, because the coefficient of extinction by the aerosol atmosphere at the wavelength of  $0.4~\mu m$  is  $0.1~km^{-1}$ . The sensitivity of this method is high as compared to other methods of chemical analysis and achieves 1 ppm.

Thus, the obtained results allow us to hope that this method is promising for quantitative analysis of the uranium content in the radioactive releases.

# 4. Method of remote real-time monitoring of $UF_6$ hydrolysis products in the atmosphere over the production area

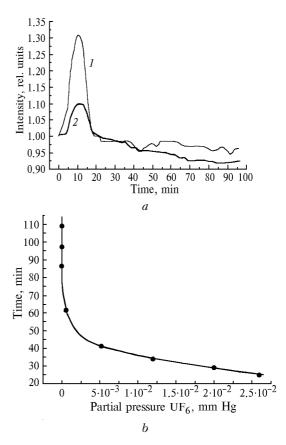
Capabilities of optical methods in real-time monitoring of uranium hexafluoride and products of its hydrolysis in the air over the production areas have been studied experimentally in the process of hydrolysis of uranium hexafluoride under controlled conditions imitating the atmosphere. The scattering and absorption coefficients were used to study the process of formation and kinetics of aerosols at interaction of UF $_6$  with water vapor.

It is known that studying the intensity of scattering in different directions it is possible to estimate the dimensions and shape of aerosol particles, as well as the rate of their growth. It should be noted that nonsphericity of particles causes significant fluctuations of scattered radiation, which can be isolated against the background of "constant" component. If a polarized sounding radiation is used, the degree of its depolarization and, consequently, the degree of particle nonsphericity of hydrolysis products is estimated. The concentration of molecular component of the hydrolysis products can be determined by spectrophotometric and photoacoustic measurements of the absorption coefficients with the time resolution  $\sim 10^{-3}$  s. Thus, the methods of optical sensing of the volume, in which hydrolysis occurs, may provide for information on the characteristics of the hydrolysis products and initial periods of the process; this information cannot be obtained by standard chemical

In the experiments,  $^{14-18}$  hydrolysis of UF<sub>6</sub> was observed at different ratios of the initial components. The ratios UF<sub>6</sub>:H<sub>2</sub>O corresponded to the cases of complete hydrolysis (1:1), stoichiometry (1:2), and excess of humidity (1:3, 1:4, 1:6, 1:8, 1:12). The initial concentration of UF<sub>6</sub> varied from 10<sup>-6</sup> to 6 mm Hg. Intensity of light ( $\lambda$ =0.6328  $\mu$ m) scattered by aerosol

was normalized to the intensity of the background radiation recorded with the detector in the beginning of measurement cycle.

Figure 4a shows the results of similar measurements at the  $UF_6$  concentration equal to 1.5 mm Hg and the water vapor concentration equal to 18 mm Hg (this value corresponds to the water vapor concentration in the actual atmosphere under standard conditions). It is seen that at tenfold excess over the H<sub>2</sub>O concentration the intense (active) volume hydrolysis of uranium hexafluoride  $^{14}$  occurs with formation of aerosols even at the low concentration of UF<sub>6</sub>, and this hydrolysis terminates within first 20 min. At lower concentrations of H<sub>2</sub>O, the process of intense hydrolysis proceeds slower and formation of aerosol has no pronounced time intervals. It should be emphasized that termination of the intense hydrolysis does not mean complete termination of the reaction, because forward and backward scattered signals do not drop down to zero level, thus indicating the presence of a "penetrative fraction" of  $UF_6$ , which continues to react with H<sub>2</sub>O and hydrolysis products.



**Fig. 4.** Dynamics of hydrolysis (a) of UF<sub>6</sub> at the UF<sub>6</sub> partial pressure of 1.5 mm Hg, and the  $\rm H_2O$  concentration of 18 mm Hg; backward (1) and forward (2) scattering; dependence of the period of active volume hydrolysis (b) on the initial pressure of UF<sub>6</sub>.

It is clear from the obtained dependences that the time of aerosol formation is mostly determined by the

time of the hydrolysis process, and the lifetime of aerosols is determined by diffusion and convective processes, as well as coagulation. At low concentrations of the parent material the effect of coagulation on the process of aerosol formation is insignificant because of the low probability of collision between particles.

No depolarization of a linearly polarized laser radiation has been observed in the experiments. The causes may be low concentration of UO2F2 particles as compared to the concentration of HF and H<sub>2</sub>O particles, small size of crystals, crystalline hydrate water, and others. Spectral analysis of oscillations of the scattered radiation did not show any pronounced maxima, which could evidence the presence of nonspherical UO<sub>2</sub>F<sub>2</sub> particles.

In the experiments, the ultimately concentrations of UF<sub>6</sub> have been achieved:  $1.2 \cdot 10^{-6}$  mm Hg at the H<sub>2</sub>O concentration equal to 6 and 12 mm Hg (Fig. 4b). Under such conditions, the ratio 1:10 is known true; this is usually the case under atmospheric conditions because of the concentration of UF<sub>6</sub> in releases (except for emergencies). The results have shown that even at such low concentrations of uranium hexafluoride close to the maximum permissible concentration (MPC), volume hydrolysis involves formation of the aerosol fraction, and the optical methods detect it with good reproducibility from one experiment to another.

As the UF<sub>6</sub> concentration decreases down to the MPC level, the period of the active hydrolysis increases almost exponentially. 18 Such a character of the dependence is explained by the decrease of the amount of nonhydrolized UF<sub>6</sub> and, as a consequence, the decrease of the probability of collision of UF<sub>6</sub> molecules with H<sub>2</sub>O molecules in the number necessary

The experiments showed that the optical methods allow rather reliable detection of low concentrations of radioactive an toxic admixtures at hydrolysis of uranium hexafluoride at the level exceeding the MPC, and the equipment based on these methods is capable of providing the reliable monitoring of pre-emergency situations; in addition, this equipment is relatively cheap and convenient in use.

Thus, the theoretical and experimental studies demonstrate that the physical grounds of indirect remote techniques for detecting radioactive contamination in the atmosphere by making use of specific traces that appear in the atmosphere due to the interactions of radioactive emissions with the atmospheric constituents open good prospect for their use in practice.

Further progress in the development of high-power lasers provides for new possibilities of using a wide class of nonlinear and coherent interactions of light and different media as a new means of laser sensing of the atmospheric parameters, first, those that may successfully be measured using traditional techniques. It is important that remote optical, including the laser

ones, and microwave methods of monitoring radio wave contamination of the environment have higher sensitivity and are less expensive. In principle, these methods allow remote studies of the radioactive contamination at different altitudes and over areas of different extents.

Although some methods of monitoring the radioactivity under emergency conditions have already been approbated, the other remote techniques from the above discussed still have only an academic significance with their practicality being strongly dependent on the possible growth of interest in ecological monitoring.

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