Investigation of relation between UV radiation flux, electric field strength, and optical-microphysical characteristics of the atmospheric boundary layer

Yu.A. Pkhalagov, I.I. Ippolitov, V.N. Uzhegov, A.V. Buldakov, and M.Yu. Arshinov

Institute of Atmospheric Optics,
Siberian Branch of the Russian Academy of Sciences, Tomsk
Institute of Optical Monitoring,
Siberian Branch of the Russian Academy of Sciences, Tomsk

Received December 29, 2001

The paper describes the tentative results of experimental investigation of the relation between the intensity of the scattered solar UV radiation, the electric field strength, the submicron component of aerosol extinction of optical radiation, and the concentration of micron-sized particles in the atmospheric boundary layer. At fine weather a significant negative correlation was observed between variations of the UV flux and the electric field strength (the correlation coefficient $p = -0.67$) and under overcast conditions this correlation broke. It was found that at the cloudless sky the micron-sized particles grow gradually under the effect of the UV solar radiation during a day, the particle concentration changed, and the size distribution broadened considerably.

Introduction

It is known that in the modern theory of climate a strong emphasis is placed on accounting for the effect of tropospheric aerosol on radiation parameters of the climatic system, especially, its albedo, the main role in whose formation is played by the submicron aerosol fraction.[1] To take the effect of interest into account, various computational and empirical aerosol models are used. Since aerosol extinction of the visible and near-infrared radiation varies significantly in time and space, it is worth estimating its effect on the climatic system using empirical models of optical parameters of the aerosol atmosphere with a few input parameters (see, for example, Ref. 2). For justified choice of input parameters in such models, it is necessary to study different physical factors contributing to the dynamics of the sought characteristic.

Formulation of the problem

As was mentioned above, the submicron fraction of aerosol particles plays the main role in formation of the albedo of the atmosphere/surface system.[1] This fraction contributes significantly to the aerosol extinction of shortwave radiation in surface haze. According to Refs. 3 and 4, the submicron aerosol in the atmosphere is generated from the ageing fine photochemical aerosol (with the particle radius $r = 0.001–0.02$ μm) produced from the gas phase under the exposure to the solar UV radiation. Hence it follows that under certain conditions the UV flux variations can result in variations of the submicron aerosol concentration and appropriate variations of aerosol extinction in the shortwave region. Since ageing fine particles grow largely due to coagulation, a key part in variations of the submicron aerosol concentration should belong to the atmospheric electric field strength. Note that in Ref. 5 a statistically significant correlation was found between the aerosol extinction of shortwave radiation and the atmospheric electric field strength.

In this connection, the goal of this work was to study thoroughly and, in particular, to measure simultaneously spectral coefficients of aerosol extinction in surface haze (in the size range from 0.44 to 1.06 μm), the disperse composition and concentration of nanometer-sized particles (from 0.003 to 0.20 μm in diameter), the atmospheric electric field strength, and the solar UV flux.

It should be noted that this work is also of interest for studying the physical reasons for the effect of solar activity variations on the earth's weather and climate. This problem arose in connection with the fact that energy variations at the atmospheric top due to solar activity variations are negligible as compared with the total solar energy and obviously insufficient for direct effect on air mass circulation or cloud formation. Hence, some mechanism should provide for significant variations in the lower atmosphere under the effect of relatively weak factors at the atmospheric top.

Now there are some hypotheses about the causal mechanisms of the relation of solar activity variations with weather and climate.[6–8] In particular, high-energy protons produced at solar flares (solar proton events) are considered to be of primary importance in Refs. 7

0235-6880/02/04 300-06 $02.00

and 8. Penetrating to the level of the troposphere in high latitudes, these protons generate a large number of ions, excited molecules, and charged aerosols, which result in formation of a cloud layer under conditions of supersaturated water vapor.

Besides the situations with solar proton events (which are rather rare), it can be assumed that under certain conditions the UV flux variations can also form a mechanism of sun–weather relations through variation of the aerosol optical thickness. This assumption is based on the well-known fact that the highest power fluctuations due to variations of the solar constant are observed just in the UV spectral range.[6] This hypothesis is also supported by experimental data of Ref. 9, where a close correlation was found between the aerosol optical thickness of the atmosphere and the Wolf (sunspot) number characterizing the solar activity.

Experiment

The experimental investigations were conducted in May 2000 at the geophysical station of the Institute of Optical Monitoring SB RAS situated in the eastern outskirts of Tomsk (56°N).

The atmospheric transmittance was measured with an automated filter photometer[10] operating in the wavelength region $\lambda = 0.44–1.06$ μm in seven spectral intervals. A SIRSh6-100 photometric lamp was used as a radiation source, and a FEU-28 photomultiplier served a radiation receiver. The measurements were conducted along a path with reflection (base length of about 430 m, total path length of about 860 m). Radiation was reflected from a mirror retroreflector (main mirror 500 mm in diameter, focal length of 1500 mm). The time of signal averaging at every wavelength was 20 s, and the time for recording of one spectrum was about five minutes. The spectral coefficients of aerosol extinction $\alpha(\lambda)$ were calculated from the recorded spectra of atmospheric transmittance using the technique described in Ref. 10. A random error of estimation of the extinction coefficient at a single measurement was 0.02–0.03 km^{-1}.

To measure the disperse composition and concentration of micron-sized particles in the atmosphere, a diffusion aerosol spectrometer operating in the size range from 3 to 250 nm was used.[11] Note that in the size range from 201 to 250 nm this spectrometer gives the net concentration of particles, which can be classified as submicron ones.

To measure the electric field strength (E), we used an automated system[12] consisting of a dynamic vibrating wire sensor of electrostatic field and a control and data processing unit. The sensor was calibrated by a 50×50×50 cm plane capacitor with edge effect compensation, to whose plates the voltage from a calibrated power supply was applied.

The UV radiation fluxes were recorded by the filter spectrophotometer[13] operating in the UV-A spectral range with the maximum effective spectral sensitivity at $\lambda = 353$ nm and 27 nm FWHM. It should be noted here that in this cycle of our investigations we measured, for some reasons, only the scattered component of UV radiation, rather than the total UV flux. However, this fact is not of principal importance, since we analyzed the diurnal variation of the UV flux, rather than its absolute values. The scattered radiation was collected from the zenith direction by quartz half-sphere from a solid angle, the plane angle at whose vertex was 110°. All the measurements were accompanied by meteorological observations.

The atmospheric transmittance was measured 24 hour a day with 3-hour periodicity. For one 30 min cycle, six atmospheric transmittance spectra were recorded, and then they were averaged at the stage of data processing (if no pronounced time trend was observed). The field strength and the UV flux intensity were measured only in daytime. Data reading was automatic with an averaging period of 1 minute. In analysis, we used both non-averaged (instantaneous) measured values of the UV flux and E (when considering the correlation between them) and the values averaged over 30 minutes for every hour (for analyzing the diurnal variation).

It should be noted that the cloudy weather prevailed in the studied region during this cycle of investigations. That is why we failed to obtain a large array of simultaneously measured parameters for slightly cloudy conditions. Nevertheless, even the tentative results of these investigations are of interest in the framework of the considered problem.

Measured results and their interpretation

For the period of measurements, a data array of about 140 averaged hourly values of the UV flux, more than 300 values of the field strength, and about 100 realizations of the averaged spectra of aerosol extinction was obtained. The measured parameters varied during the measurement period within the following limits: air temperature (t) from −2°C to +26°C, relative humidity (RH) from 25 to 100%, electric field strength (E) from −8 to 270 V/m, intensity of scattered UV flux (Φ_A) from 0.3 to 3 mW/(m^2·nm), the aerosol extinction coefficient $\alpha(0.56)$ from 0.04 to 0.28 km^{-1}, what corresponds to variation of the meteorological range from 97 to 14 km. The mean values of these parameters and their rms deviations (RMSD) over the measurement period are given in the Table. The electric field and UV radiation were measured practically every minute during 24 hours, so it was interesting to consider their mutual variability under various weather conditions.

Table. Mean values and rms deviations (RMSD) of parameters: $\alpha(0.56)$, RH, t, E and Φ_A for the data array

Statistic parameter	Measured parameter				
	$\alpha(0.56)$, km^{-1}	RH, %	t, °C	E, V/m	Φ_A, mW/(m^2·nm)
Mean value	0.138	59	11.15	114.16	1.325
RMSD	±0.046	± 23.26	± 7.01	± 48.48	± 0.714

Figure 1 shows the correlation between instantaneous values of the intensity of scattered UV radiation and the electric field strength obtained in slightly cloudy day (Fig. 1*a*) and under overcast conditions (Fig. 1*b*).

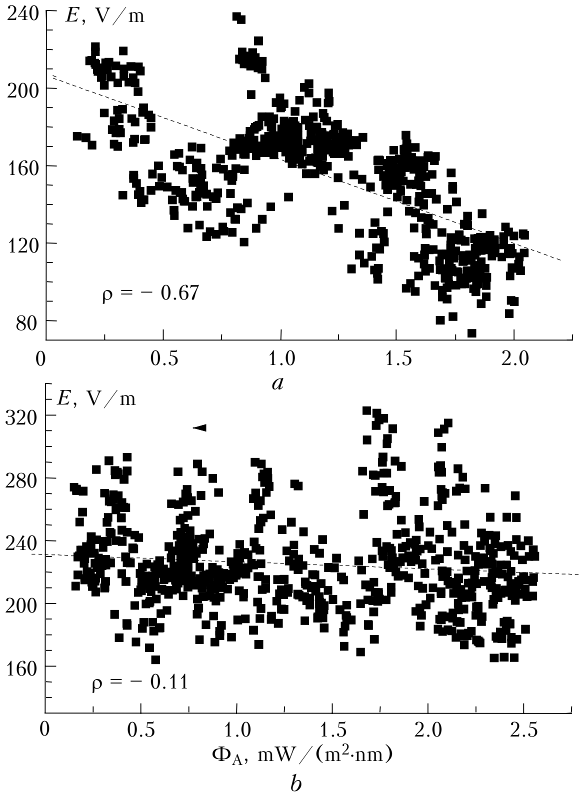

Fig. 1. Correlation between the field strength and the scattered UV radiation intensity in slightly cloudy day (*a*) and under overcast conditions (*b*), ρ is the correlation coefficient.

As is seen from Fig. 1, at slightly cloudy weather a significant negative correlation exists between these parameters (the correlation coefficient $p = -67$), i.e., the electric field strength decreases with the increasing UV flux. This indicates that at fine weather the solar UV radiation affects, in some way, the air conductance and, hence, the number of light ions in the atmosphere.[14] Under overcast conditions this correlation breaks almost completely.

The negative correlation between the parameters Φ_A and E under the conditions of slightly cloudy weather can be also seen from the character of diurnal variability of the average values of the scattered UV radiation intensity and the electric field strength dated to May 26 and depicted in Fig. 2.

As is seen from Fig. 2, the field strength decreases considerably in the morning with the increasing UV flux, then two minima are observed in the afternoon (1 p.m. and 6 p.m.), and in the evening (after 6 p.m.) the field strength increases again.

Analysis of the available literature on atmospheric electricity shows that the character of the diurnal variability of the electric field strength revealed in this work agrees with the diurnal variation of E obtained earlier for continental stations.[15] It is characterized by the high value of the field strength in the morning, the low value in the afternoon, and the increase in the evening.

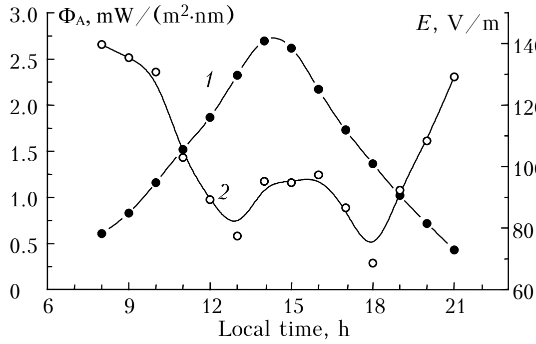

Fig. 2. Diurnal variation of the scattered UV radiation intensity (*1*) and the atmospheric electric field strength (*2*) at slightly cloudy weather (May 26).

Hence, based on comparison of the diurnal variations of the field strength and the UV radiation flux, we can assume that a source of light nanometer-sized ions connected with UV radiation operates in daytime. It is known[16] that light nanometer-sized ions make the major contribution to the air conductance. The UV radiation at $\lambda > 300$ nm itself cannot produce ionization in a molecular medium, since the ionization potentials of major and minor atmospheric molecules are in the shorter-wave UV range, but the radiation of this range does not reach the Earth's surface because of ozone absorption.

It is known that the radiation of the UV-B range ($280 < \lambda < 315$ nm) reaches the troposphere and significantly affects photochemical processes, which are especially active when anthropogenic emissions, nitrogen oxides, sulfur oxides, and various organic compounds are present in the troposphere. As a result, the secondary gas pollutants are produced,[17] which generate aerosol particles in the presence of organic compounds.[18,19]

As was shown in Ref. 20, the solar UV radiation in the urban atmosphere leads to simultaneous transformation of the size and composition of all aerosol fractions due to heterogeneous photochemical reactions because of accumulation of organic material on particles. This process proceeds in both fair and overcast days, and its intensity strongly depends on the solar radiation intensity. As a result, not only the particle size spectrum deforms, but the particle absorption spectra in the UV range change too. In Refs. 20 and 21, formation of fine (r_{av} ~ 50 nm) aerosol was observed when a purified air flow containing aldehyde admixtures was exposed to UV radiation of a mercury-vapor lamp. In that case, the aerosol particle number density measured with an aerosol counter was about 10^6 cm^{-3}. In the case of benzaldehyde admixture, the produced aerosol matter could absorb radiation in the range from 306 to 328 nm.

According to Ref. 22, phenol–$(H_2O)_n$ complexes with n varying from 1 to 12 show the tendency to the decrease of the ionization potential. Possibly, clusters with the ionization potential less than 4.13 eV (λ >300 nm) are produced in the process of complex formation. Just organic admixtures in homogeneous and heterogeneous reactions can likely be considered as ionization agents under the exposure to either UV radiation or natural soil and air radioactivity.

In what follows, we consider peculiarities of diurnal variations of various average parameters obtained in this experiment. Besides the UV radiation flux Φ_A and the electric field strength E, the analyzed parameters include aerosol extinction of optical radiation largely due to the submicron aerosol fraction $\Delta_\alpha(0.44) = \alpha(0.44) - \alpha(1.06)$, the concentration of micron-sized aerosol N_{ms} (in the range from 3 to 200 nm), and the relative air humidity RH.

It turned out that the character of the average diurnal variation of the scattered UV radiation under overcast conditions did not differ from that at slightly cloudy weather, while the diurnal variation of the field strength can vary significantly. Figure 3 shows the diurnal variations of the averaged values of Φ_A and E, as well as $\Delta_\alpha(0.44)$, N_{ms}, and RH for May 24 under overcast conditions.

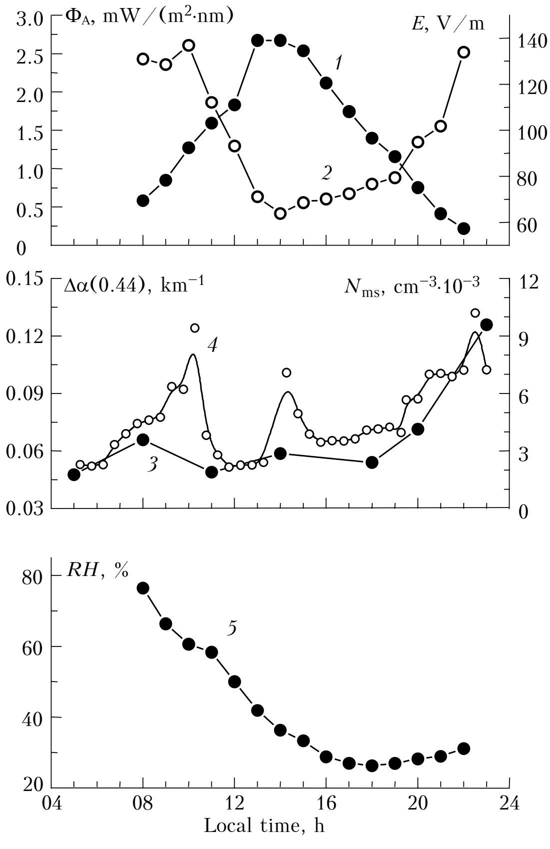

Fig. 3. Diurnal variation of the scattered UV radiation intensity (curve *1*), electric field strength (*2*), submicron component of aerosol extinction (*3*), concentration of micron-sized aerosol particles (*4*), and relative humidity of air (*5*) on May 24 under overcast conditions.

It can be seen that the diurnal variation of the electric field here had only one minimum (at 2 p.m.). Nevertheless, the negative correlation remains between Φ_A and E. Variations of the parameter $\Delta\alpha(0.44)$ in the first half of a day were only slightly pronounced, and after 6 p.m. it increased significantly.

The concentration dynamics of micron-sized particles during a day is rather pronounced: from 2000 cm^{-3} (in the morning) to 10000 cm^{-3} (in the evening) with a maximum at 10 a.m. in the active part of increasing UV flux intensity. To be noticed is a certain similarity of tendencies in the diurnal variations of the submicron component of aerosol extinction and the concentration of micron-sized particles. An unambiguous physical interpretation can hardly be given to this fact, since particles of several tens of nanometers in size cannot be directly (through scattering) observed in the visible spectral region. Another noteworthy feature is the coordinated variation of $\Delta\alpha(0.44)$ and E in the evening.

Under the conditions of broken clouds (Fig. 4) the diurnal variation of the electric field strength had two maxima at 11 a.m. and 8 p.m. and one minimum at 2 p.m.

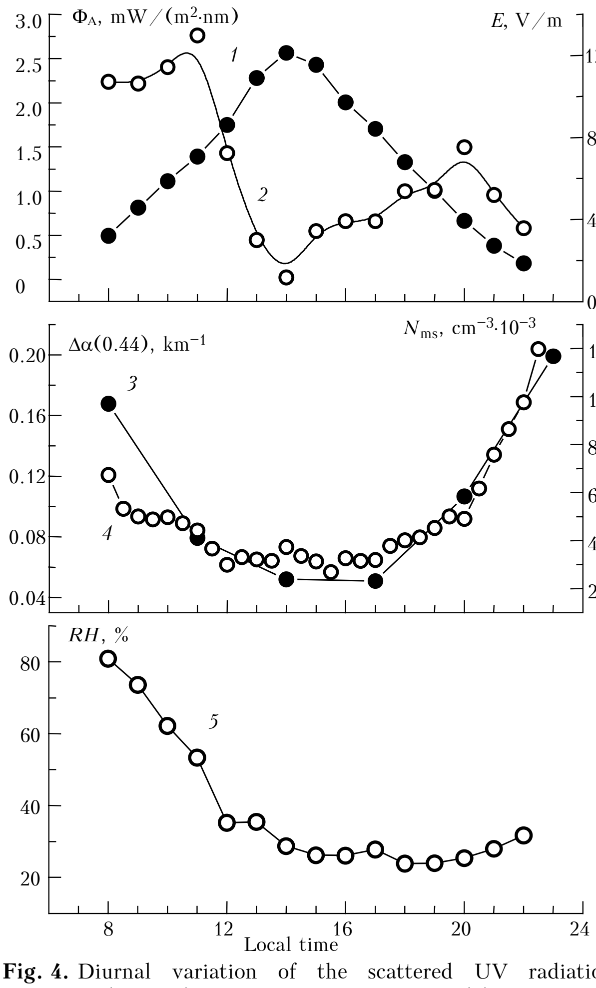

Fig. 4. Diurnal variation of the scattered UV radiation intensity (curve *1*), electric field strength (*2*), submicron component of aerosol extinction (*3*), concentration of micron-sized aerosol (*4*), and the relative air humidity (*5*) at broken cloudiness (May 21, cloud amount from 3 to 10).

The diurnal variation of the submicron component of aerosol extinction $\Delta\alpha(0.44)$ is characterized by a pronounced wide minimum at 3–4 p.m., what poorly corresponds to the diurnal variation of the electric field strength. It is characteristic that the diurnal variation of the micron-sized particle concentration in this day, as well as in the overcast day, faithfully copies the diurnal variation of aerosol extinction in the shortwave spectral region.

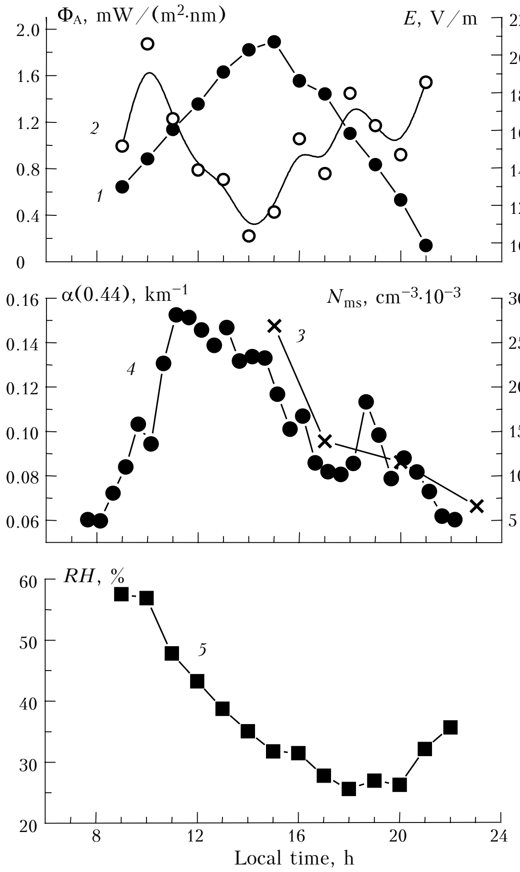

Fig. 5. Diurnal behavior of the scattered UV radiation intensity (curve *1*), electric field strength (*2*), submicron component of aerosol extinction (*3*), concentration of micron-sized aerosol (*4*), and the relative air humidity (*5*) at cloudless sky (May 1).

Finally, Fig. 5 shows the diurnal variations of the studied parameters for a cloudless day (May 1). It can be seen that the electric field strength here had the morning and evening maxima and one minimum (at 2 p.m.). The comparison of the diurnal variations of the micron-sized particle concentration and the aerosol extinction coefficient $\alpha(0.44)$ for the period from 3 p.m. to 11 p.m. (measurements of aerosol extinction in this day were started only at 3 p.m.), as well as in the previous figures, points to the common tendencies in their variability. However, the most pronounced peculiarity of this day is the presence of the marked positive correlation between the UV flux intensity and the micron-sized aerosol concentration. This correlation manifests itself against the background of rather low relative air humidity (< 60%). This result seems to be very important from the viewpoint of the above hypothesis about the influence of the solar UV radiation on generation of submicron aerosol.

In this connection, it is interesting to follow the time dynamics of the particle size spectrum and the concentration of micron-sized particles depicted in Fig. 6 for the period from 10 a.m. to 4 p.m.

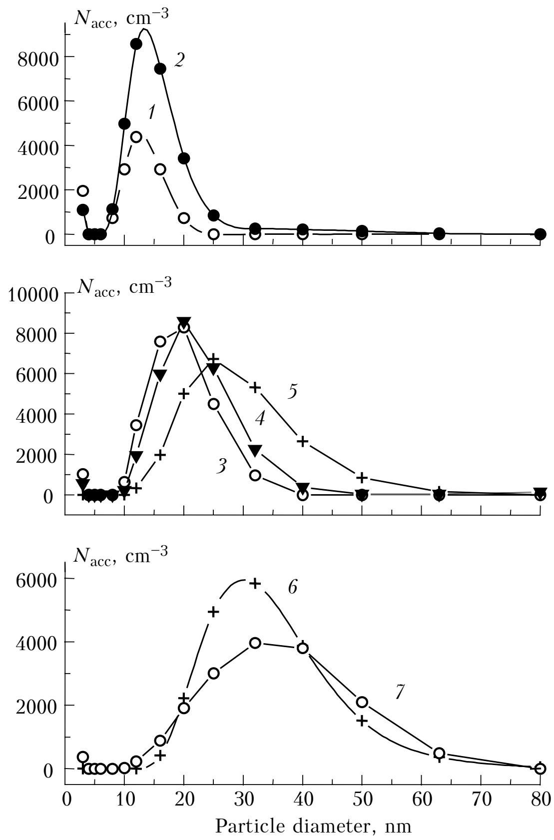

Fig. 6. Diurnal dynamics of the particle size spectrum and concentration of micron-sized particles at cloudless sky (May 1). Curves *1*, *2*, *3*, *4*, *5*, *6*, and *7* were obtained at 10, 11, 12 a.m., 1, 2, 3, and 4 p.m., respectively.

It follows from the above data that at 10 a.m. the particle fraction with the maximum (in diameter) in the range from 12 to 13 nm and with the concentration of about 4000 cm^{-3} was formed in the atmosphere. At 11 a.m. the distribution maximum shifted to 15 nm, and the particle concentration increased up to 9000 cm^{-3}. At 12 a.m. the distribution maximum was already at 20 nm, but the particle concentration in this case decreased down to 8000 cm^{-3}. At 1 p.m. a slight particle growth continued at the unchanged concentration, and at 2 p.m. the particle size spectrum broadened noticeably with the maximum shifted to 25 nm and the concentration decreased down to 6000 cm^{-3}. At 3 and 4 p.m. the particle growth continued with the simultaneous broadening of the particle size spectrum and decrease of the concentration. Thus, by 4 p.m. the particle distribution peaked already at 35–40 nm at the concentration less than 4000 cm^{-3}.

The found regularities in the diurnal variation of the size spectrum and the concentration of micron-sized particles under the conditions of cloudless weather are obviously indicative of the effect of solar UV radiation on the process of atmospheric aerosol generation.

Conclusion

Based on the analysis of simultaneously measured spectral coefficients of aerosol extinction in surface haze, the disperse composition and concentration of nanometer-sized particles, the atmospheric electric field strength, and the intensity of the solar UV radiation flux ($\lambda = 353$ nm), it was found that at the slightly cloudy weather the electric field strength decreased with the increasing UV flux. The correlation coefficient between these parameters was $p = -0.67$. It was assumed that light ions of nanometer size are produced during a day under the exposure to the solar UV radiation in the presence of organic pollutants in the troposphere. These ions make the major contribution to the air conductance. It was suggested that the ionization potential of particles decreases considerably due to accumulation of organic material on them.

In a cloudless day a well pronounced positive correlation was observed between the UV flux intensity and the concentration of micron-sized aerosol. The dynamics of size spectrum and the concentration of micron-sized particles was analyzed. It was found that aerosol particles increased gradually during a day under the exposure to the UV radiation, the particle concentration changed, and the size distribution broadened.

As a whole, the obtained results have shown that the questions about the possible effect of the solar UV radiation and the electric field strength on the optical characteristics of the aerosol atmosphere are valid, although they were formulated speculatively.

Acknowledgments

This work was partly supported by the Russian Foundation for Basic Research (Grants No. 99–5–64564A and No. 01–05–65197).

References

1. T.A. Tarasova and E.M. Feigelson, Izv. Akad. Nauk SSSR, Ser. Fiz. Atmos. Okeana **18**, No. 11, 1199–1206 (1982).
2. M.V. Panchenko, S.A. Terpugova, and V.V. Pol'kin, Atmos. Oceanic Opt. **11**, No. 6, 532–539 (1998).
3. G.V. Rozenberg, G.I. Gorchakov, Yu.S. Georgievskii, and Yu.S. Lyubovtseva, in: *Physics of the Atmosphere and Problems of Climate* (Nauka, Moscow, 1980), pp. 216–257.
4. G.V. Rozenberg, Izv. Akad. Nauk SSSR, Ser. Fiz. Atmos. Okeana **18**, No. 11, 1192–1198 (1982).
5. Yu.A. Pkhalagov, V.N. Uzhegov, E.V. Ovcharenko, V.N. Genin, V.A. Donchenko, M.V. Kabanov, and N.N. Shchelkanov, Atmos. Oceanic Opt. **12**, No. 2, 99–102 (1999).
6. B.M. McCormack and T.A. Seliga, eds., *Solar-Terrestrial Influences on Weather and Climate* (D. Reidel, Dordrecht, 1979).
7. L.S. Ivlev and S.N. Khvorostovskii, Atmos. Oceanic Opt. **13**, No. 12, 993–999 (2000).
8. L.S. Ivlev and S.N. Khvorostovskii, Atmos. Oceanic Opt. **13**, No. 12, 1000–1004 (2000).
9. V.K. Roldugin and G.V. Starkov, Dokl. Akad. Nauk SSSR **370**, No. 5, 675–677 (2000).
10. Yu.A. Pkhalagov, V.N. Uzhegov, and N.N. Shchelkanov, Atmos. Oceanic Opt. **5**, No. 6, 423–427 (1992).
11. M.Yu. Arshinov, B.D. Belan, V.K. Kovalevskii, et al., Atmos. Oceanic Opt. **13**, Nos. 6–7, 580–583 (2000).
12. V.A. Donchenko, M.V. Kabanov, V.T. Kalaida, et al., Izv. Vyssh. Uchebn. Zaved., Ser. Fizika, No. 5, 88–98 (1998).
13. A.A. Eliseev, I.I. Ippolitov, M.V. Kabanov, et al., Atmos. Oceanic Opt. **7**, No. 5, 301–302 (1994).
14. V.V. Smirnov, *Electric Factors of Air Purity*, Trudy Ins. Exp. Meteorol., Is. 30(104), 64–106 (1983).
15. I.M. Imyanitov and K.S. Shifrin, Usp. Fiz. Nauk **LXXVI**, No. 4, 593–642 (1962).
16. V.V. Smirnov, *Ionization of the Troposphere* (Gidrometeoizdat, St. Petersburg, 1992), 311 pp.
17. I.G. Garvert, in: *The Chemistry of the Polluted Troposphere, Proc. NATO. Adv. Study Just. Island of Corfu,* Sept. 28–Oct. 10 (1981) (Dordrecht, 1982), pp. 425–456.
18. L.S. Ivlev, in: *Problems of Atmospheric Physics*, (SPbGU Press, 1997), Is. 20, pp. 54–80.
19. R.A. Duce, V.A. Mohnen, P.R. Zimmerman, et al., Reviews of Geophysics and Space Physics **21**, No. 4, 921–952 (1983).
20. G.I. Skubnevskaya and G.G. Dul'tseva, Atmos. Oceanic Opt. **10**, No. 6, 433–438 (1997).
21. G.I. Skubnevskaya, G.G. Dultseva, and S.N. Dubov, Atmos. Oceanic Opt. **13**, Nos. 6–7, 562–564 (2000).
22. K. Kleinermanns, C. Janzen, and D. Spanbenberg, J. Phys. Chem. A **103**, No. 27, 5232–5239 (1999).