Active spectral nephelometry in the study of microphysical characteristics of submicron aerosol

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The approach to investigation of the properties of submicron atmospheric aerosol called "the method of active spectral nephelometry" is described in this paper. The technique and results of spectral polarimetric investigation of submicron aerosol with controlled influence on its properties are discussed in detail. Measurements were carried out by means of the instrumentation complex including a FAN nephelometer, the devices for humidification and heating of aerosol, and the block for cut-off of fine particles using a 4-stage diffusion battery. Solution of the inverse problem provided for obtaining the data on the size spectrum and the refractive index of aerosol particles. The estimates of the size dependence of hygroscopicity and volatility of aerosol particles are obtained. Measurements with the use of the diffusion cutter confirmed its calculated calibration function and the lower boundary of the retrieved size spectrum.

Introduction

Atmospheric aerosol is one of the most variable components of the Earth's atmosphere. Adequacy of the climate and radiative models is to a great degree determined by the reliability of data on the aerosol optical and microphysical properties.

Great variety of sources and sinks, chemical composition of atmospheric aerosols, and high spatiotemporal variability of their properties make the idea that at present the most reliable data on the aerosol optical characteristics and their transformation under the effect of the whole complex of geophysical processes can be obtained only experimentally.

In this paper, we discuss the approach to investigation of the submicron aerosol properties in the atmosphere, which we have been developing during many years, and conventionally call it "the method of active spectral nephelometry."

1. Principles of the method

Before describing the technique, let us note the following key features of the approach developed.

It is obvious that a comprehensive study of the aerosol properties is only possible if a complex of various methods and tools are used for measuring the aerosol microstructure parameters, its chemical composition, particle shapes, and the spectral dependence of the complex refractive index. In order to reveal the relations of all these characteristics to the external geophysical factors, the long-term wide-scale experiment is needed. However, even in the case when the experiment has been carried out in real atmosphere and all necessary parameters have been recorded, it is hard to correctly describe many processes, because all the processes occurring in the atmosphere are in complicated relations between each other.

In particular, let us consider the well-known example, when the attempt is undertaken to parameterize the relation of optical or microphysical characteristics to the relative humidity of the air using data obtained from *in situ* measurements. Let us emphasize that it is one of the very important processes determining the variability of the aerosol optical properties, the account of which is urgently needed.[1] The relative humidity of the air changes in the near-ground layer of the atmosphere during a day synchronously with the change of air temperature. Transformation of the aerosol microstructure and the change of its concentration under the effect of generation, transfer, emission, and sedimentation of particles occur simultaneously with this well pronounced process. It is clear that combined analysis of all these processes is necessary for a correct description of the effect of relative humidity of the air. Otherwise, if only the data of observation of any aerosol characteristic and the value of relative humidity are used for the estimation, it is practically impossible to obtain the correct relation sought.

In order to overcome these unavoidable problems, the variability of the dry matter of aerosol particles under the effect of external factors and the aerosol condensation activity were studied separately. This separation principle made the basis of the "active spectral nephelometry" method we have developed.

To achieve this task we combine observations of the dry matter of aerosol particles in a monitoring mode and regular measurements of the dependence of aerosol properties on the relative humidity of air under controlled influence.[2] The additional way of active influence, which was also put into practice of daily measurements, is the so-called thermooptical

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method. This method allows us to qualitatively estimate the relative content of the substances of different volatility in the atmospheric aerosol[3] by means of controlled heating of atmospheric aerosol particles from ambient air temperature up to $T \sim 250°C$.

Thus, the first key item of the approach is combination of the monitoring regime of measurements of the atmospheric aerosol characteristics with the controlled influence, which makes it possible to extend the information content of the experiment. The usefulness of such an approach has been confirmed by many-year practice (see, for example, Refs. 2, 4, 5) regardless that the measurements of the scattering coefficients have been conducted only at a single wavelength.

The next stage in the development of this method is related to passing to measuring the coefficients of directed scattering, at the angle of 45° at three wavelengths (0.41, 0.5, and 0.63 μm) and at 90° scattering angle using polarized and cross polarized components of the scattered light at two wavelengths (0.44 and 0.51 μm).

It is the principle moment here that in this case it is possible to solve the inverse problem that provides for obtaining data on the size spectrum and the refractive index of the aerosol particles.

Let us note the following, in our opinion, important circumstance, which emphasize the prospects of such an approach to estimation of the optical characteristics. Obviously, to solve the radiative problems, it is necessary to know different aerosol optical parameters in different wavelength regions, in addition to that, the measurements are being carried out and sufficient statistics is provided. Nowadays a wide variety of methods and means are used for studying the aerosol properties in the atmosphere.[6] One can select two approaches to estimation of the optical characteristics of aerosols.[6] One of them makes use of the data on microphysical and chemical composition of particles with the subsequent calculation of the optical characteristics. The other one uses the results of optical measurements.

The advantage of the "microphysical" approach is the possibility of obtaining, by calculations, the data on practically all necessary aerosol optical parameters in any wavelength region. However, in our opinion, it is only an illusive advantage. The most serious disadvantage of this approach is the fact that any restriction or distortion of the data on the microphysical parameters of particles or on their shape can lead to the uncontrollable errors in retrieving the optical image that can hardly be estimated. Especially unpredictable errors in estimating the optical parameters of the aerosol appear in trying to calculate the complex refractive index of particles by use of data on chemical analysis of the aerosol matter.

The approach to description of the optical properties of atmospheric aerosol based on the results of investigations of the optical characteristics directly in the real atmosphere is substantially free of disadvantages of the microphysical modeling. One can consider as a disadvantage the fact that the use of data obtained in this case are, as a rule, restricted to the range of optical characteristics and the wavelength region, where the initial observation data were obtained, and going out of their frameworks requires additional investigations.

The approach, which G.V. Rosenberg called "the method of microphysical extrapolation"[7–11] allows one to partially overcome these difficulties. It is based on the idea of using the data on the microstructure and the complex refractive index obtained from inversion for subsequent calculation of the optical characteristics (which were not directly measured in the experiment).

Based on our experience, it is necessary to emphasize that one should very conscientiously use the results of retrieval of the microstructure and the refractive index from the data of measurements of the small number of optical parameters. Without some special experiments those can hardly be used for the detailed study of the behavior of the particle size-distribution function. At the same time, these data are most suitable for the subsequent calculation of the optical characteristics. As concerning the retrieval of other optical characteristics at the same wavelengths or in the wavelength region close to it, the optical properties are essentially determined by the same range of the particle size. It is statistically manifested as high correlation between the optical parameters. Nevertheless, the limited wavelength region and small number of the measured characteristics (as in our case), which are used in solving the inverse problem, require careful study and correct determination of both the boundaries of the retrieved size spectrum, and the errors in estimating the refractive index.

Thus, the main idea of the method of active spectral nephelometry is in combining routine optical observations, controlled influence on the aerosol, and the corresponding apparatus for solving the inverse problems.

Obviously, such an approach can be realized by use of different optical devices with the localized measurement volume.

2. Realization of the method for studying the submicron aerosol

Having in mind that the instrumentation, techniques for calibration and measurements have been quite thoroughly described in our previous publications,[2,12] here we would like to discuss only the key moments necessary for the further description.

The optical instrument, by means of which all the measurements discussed have been carried out, was a FAN nephelometer equipped with the devices for producing active effects on the aerosols. This setup is a part of the IAO SB RAS station for *in situ* monitoring of the aerosol (http://aerosol1.iao.ru). The system provides for hourly measurements in the automated mode. Thus, when realizing the active regime of influence, the second identical nephelometer, aethalometer, photoelectric counter of particles and meteocomplex of the aerosol station allow one follow up the variations of the atmospheric situation.

One more, also very important moment, is that all the schemes of organization of continuous pumping of atmospheric aerosol through small volumes of nephelometers certainly restrict the capabilities of investigation of particles, the radius of which exceeds 1 μm. The fact that measurements are carried out in the wavelength range 0.41 to 0.63 μm hampers obtaining the data on the larger particles.

It is for this reason that in the title of the paper and in discussion of the results we mention only submicron particles.

2.1. Technique for inverting the data of optical measurements

The inverse problem was solved by the iteration method[13] based on the Twitty algorithm.[14] The advantages of the iteration method are the simplicity of its realization on a computer and an automated possibility of providing for a non-negative solution. However, the iteration algorithm does not yield the estimates of the errors of retrieval. Estimation of the reliability of a solution of the inverse problem and the range of the retrieved size spectrum was carried out by means of the numerical experiments (the lower boundary was also determined in the experiments based on the controlled cut-off, which will be described below). Adding random errors corresponding to the measurement errors simulated the noise of the modeled and measured optical characteristics. As is shown in Fig. 1, the distributions obtained at a large number of realizations of random disturbances, fill in the corridor between two envelopes. This gives an idea of how to estimate the reliability of the solution of the inverse problem. Comparison of different model distributions with the results of inverting their optical images has shown that the size spectrum is reliably retrieved in the size range from 0.07 to 0.6–0.8 μm.

One can visually get an idea of the range of sensitivity of the parameters measured by use of a FAN nephelometer to the particles of different size, if calculating the contribution of the particles to the observed optical characteristics depending on their size. Such dependences obtained by use of a single-parameter model of the atmospheric haze[10,15,16] at different values of the visibility range are shown in Fig. 2.

When inverting the spectral nephelometric data, the refractive index and, hence, the kernel of the Fredholm equation are assumed to be *a priori* unknown. Numerical experiments with a model particle size-distribution have shown that the norm of the second derivative of the logarithm of the size spectrum can serve a criterion for the choice of the refractive index. Besides, it became clear that it is impossible to determine the real n and imaginary κ parts of the refractive index separately for the set of the optical parameters measured with a FAN nephelometer having in mind the accuracy of measurements realized in the experiment. The optical discrepancy and the smoothness of the solution practically do not change at such correlated change of n and κ that their difference is constant. Let us note that such an ambiguity does not affect the shape of the retrieved particle size distribution. When inverting the measurement data, the rms deviation of the retrieved optical parameters does not exceed 5% in the majority of cases.

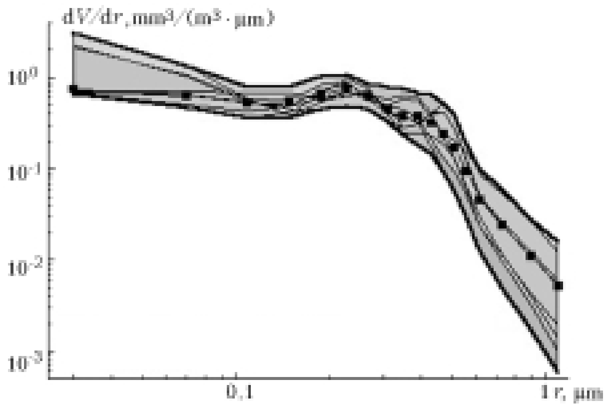


Fig. 1. The results obtained by inverting the measured and "disturbed" optical parameters: squares present the results of inversion of the measured optical parameters, thin lines are the inversion of the optical parameters with the errors, solid lines are the envelopes.

Interpretation of the data of optical measurements aimed at estimation of the microphysical parameters is a sort of the so-called ill-posed problems. Therefore, any *a priori* data available on the properties of the medium (for example, on the shape of the particle size distribution or some its parameters, on the value of the complex refractive index) or, at least, the knowledge of the reasonable boundaries of their variability could allow one to overcome the difficulties in seeking the true solution.

As was mentioned above, the data on the size spectrum obtained from the solution of the optical inverse problem, as a rule, are yet insufficient for estimating all the peculiarities in the particle size-distribution function. In order to overcome this disadvantage, we have developed a method for artificially limiting the size range of particles by means of cut-off procedures of different types. Ideally, such an approach, which is the natural development of the "active nephelometry," allows one to measure the optical characteristics of the quasi-monodisperse ensemble of particles. Subsequent solution of the inverse problem in this case makes the basis for a detailed study of the particle size-distribution function and estimation of the refractive index as a function of particle size.

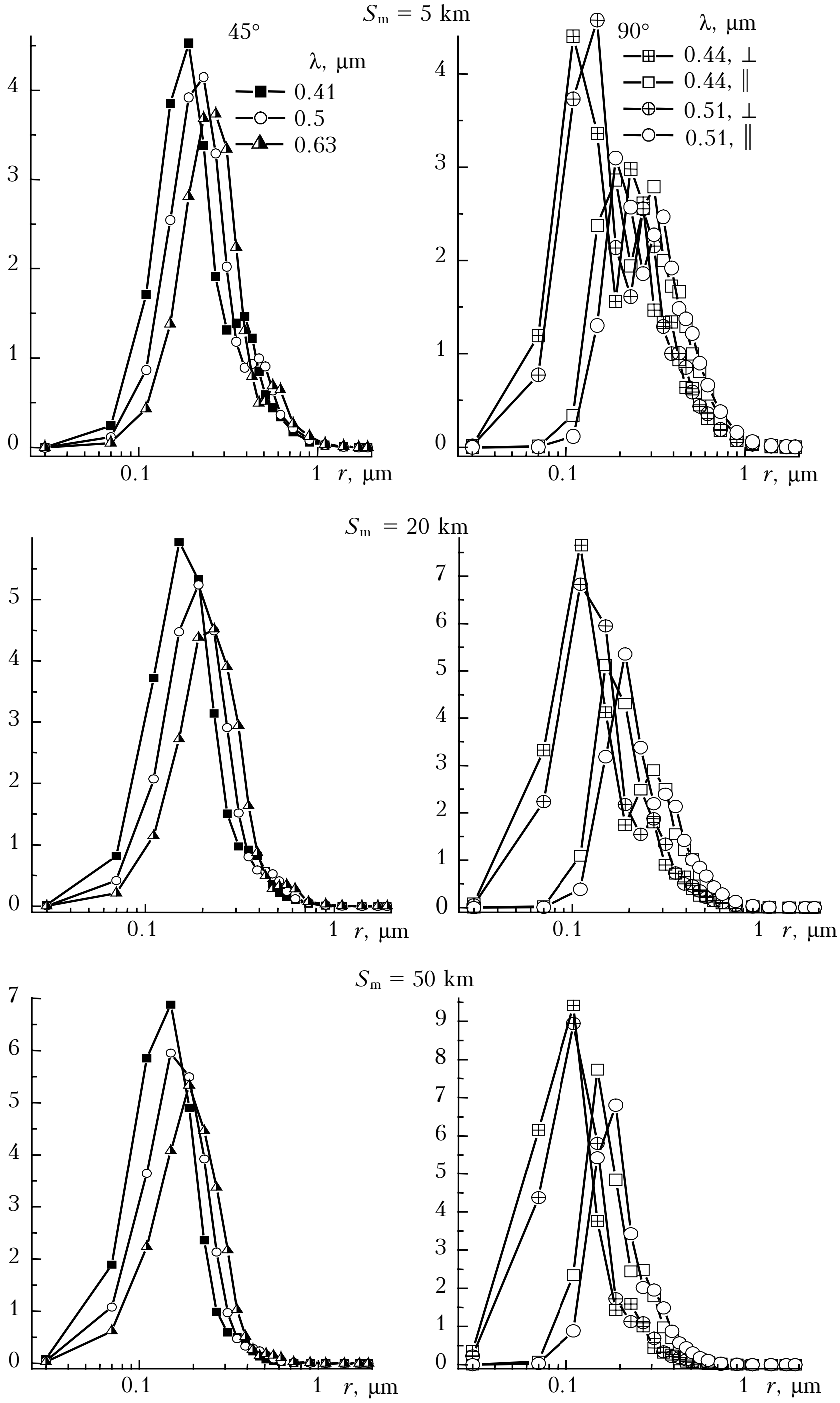


Fig. 2. Contribution of particles of different size to the optical characteristics measured.

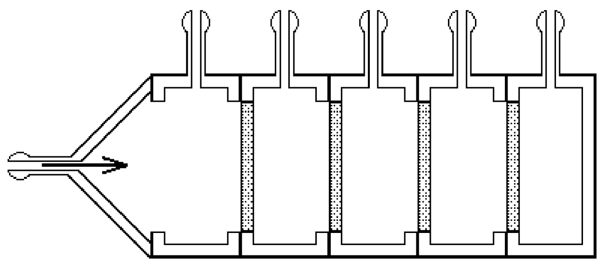
At present, we have succeeded in realizing and developing the technique for a controlled restriction of the size range of the particles measured only from the side of their lower boundary (four successive cascades of a diffusion battery). The attempt of using the cut-off from the upper boundary of the size spectrum using the device based on the corona discharge did not lead to the desirable result yet. However, even at the present stage of the development of the method the use of a successive cut-off allows one, on the one hand, to estimate the sensitivity of the method in solving the inverse problem and, on the other hand, to use these data as *a priori* and then to increase the quality of inversion.

2.2. The cut-off method

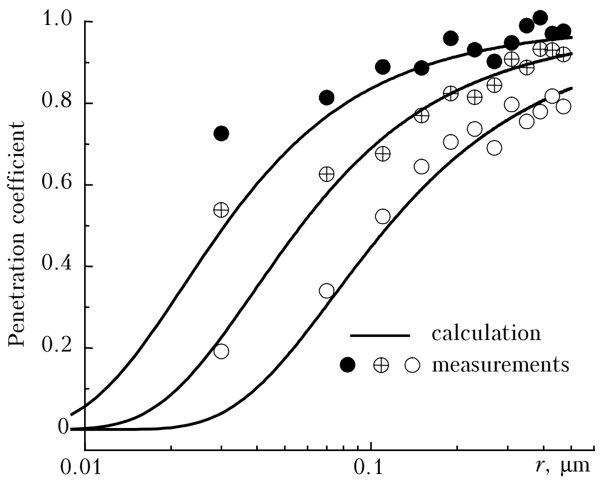
The instrumental realization of the diffusion cutter and its detailed laboratory calibration has been carried out at the Institute of Chemical Kinetics and

Combustion SB RAS.[17] In parallel this device has also been adapted to the nephelometer system, and first measurements of aerosol were carried out.[18,19]

The block-diagram of the diffuse cutter is shown in Fig. 3. Its operation principle is analogous to that of the diffusion battery[20] and is based on the well-known dependence of the diffusion coefficient of particles on their size.



a



b

Fig. 3. Block-diagram of the diffusion cutter (*a*) and the penetration coefficients of its different stages (*b*) (the results calculated according to Refs. 16 and 20 and retrieval from the measurement results).

The diffusion cutter removes the smallest particles from the airflow. As the number of corresponding screens increases, the boundary of the cutting moves toward larger size. The change of the number of cascades combined with the selection of the corresponding screens provides for a controlled change of the lower boundary of the particle size-distribution function in the airflow under study.

2.3. Approbation of the measurement technique using the cut-off and solution of the inverse problem

The development of the technique for measurements by means of the diffusion cut-off was carried out in the series of experiments at the IAO aerosol station[18,19] in 2002–2003. Taking into account specific character of the spectral nephelometric approach, two problems were simultaneously studied in these experiments. On the one hand, measurements of the optical characteristics and subsequent solution of the inverse problem provided for retrieving size spectra and estimating the efficiency of the effect of the cut-off on this basis. On the other hand, comparison of the inversion results with the data of direct calculations of the efficiency of the cascades of the diffusion battery[16,20] allowed us to additionally examine the quality of the solution of the inverse problem and to estimate the lower boundary of the size spectrum retrieved.

The data on the penetration coefficient of different stages of the cutter are also shown in Fig. 3 (solid curves show the results calculated according to Refs. 16 and 20; the symbols show the results of corresponding solution of the inverse problem using the data of direct optical measurements).

As is seen, good agreement is observed between the calculated and retrieved data in the radius range from 0.07 to 0.4 μm.

Then one can draw the following conclusions:

1. The effect of the diffusion cutter provides for a controlled setting of the lower boundary of the aerosol particle size spectrum according to calculated characteristics (it can be used as *a priori* extra posing of the inverse problem).

2. The set of the optical characteristics and the wavelength range within which measurements were carried out, certainly allows us to determine the shape of the size-distribution function of the particles with the radius larger than 0.07 μm, but it is hardly suitable for the studying smaller particles.

3. Approbation of the method for the study of the growth factor of particles

As has already been mentioned above, a change in the relative humidity of air leads, as a rule, to a change in the size spectrum and the values of the refractive index of particles.[6,21] In our case the study of the aerosol condensation activity is being done by measuring the optical characteristics of aerosol sampled from the atmosphere, when particles undergo artificial humidification in the range of relative humidity values from 20 to 95%.

Optical characteristics of the submicron aerosol fraction as functions of relative humidity of the air are satisfactorily described by the Kasten empiric formula[21–24]:

$$\mu(RH) = \mu(RH = 0)(1 - RH)^{-\gamma}, \qquad (1)$$

where μ is the scattering coefficient or the directed scattering coefficient at the angle of 45°; RH is the relative humidity; γ is the parameter of the condensation activity. Assuming that the aerosol condensation activity does not depend on its size, one can estimate the parameter χ characterizing the particle growth as relative humidity increases, using the value of the parameter γ. Such an estimate is quite rough, because it requires setting the refractive index of the aerosol dry matter and the model of the particle size distribution. Nevertheless, its accuracy is enough for monitoring the aerosol condensation activity, as well as for analysis of the peculiarities of its temporal variability and relations to the meteorological parameters of the atmosphere.

To study the condensation transformation of the aerosol size spectrum, in particular, the dependence of the growth factor on the particle size, it is necessary to measure the maximum possible number of the optical characteristics and to use the technique for solving the inverse problems of light scattering.

In our experiments, the measurements have been carried out successively at three wavelengths. One measurement cycle lasted during about 40 minutes. Stability of the situation was followed up by additional nephelometer, which continuously recorded the directed scattering coefficient at the angle of 45° at the wavelength of 0.51 μm. An example of one of the realizations of the measured optical characteristics during humidification is shown in Fig. 4.

Transformation of the particle volume size distribution is shown in Fig. 5*a*. Correlated increase of the volume concentration of particles and the modal radius of the size distribution as relative humidity increases is well seen here. Figure 5*b* shows how the refractive index and the total volume of particles change.

Solution of the inverse problem at some stages of the artificial condensation process makes it possible to estimate the dependence of the condensation activity on the size of dry particles. This dependence for the particles larger than 0.1 μm, when the effect related to the curvature of the particle surface have become inessential, is determined by the chemical composition of aerosol and, first of all, by the content of soluble species. Such parameter as the growth factor is used for description of the condensation activity of particles of different size:

$$GF = \frac{r(RH = 80\%)}{r(RH = 30\%)}. \qquad (2)$$

The dependence of the condensation growth factor on the particle size was estimated by means of a comparison of the integral number densities:

$$N(r_j) = \int_{r_j}^{\infty} \frac{\mathrm{d}N}{\mathrm{d}r}\mathrm{d}r. \qquad (3)$$

Let us denote by N_d and N_w the distributions corresponding to dry and wet aerosol. If $N_\mathrm{d}(r_1) = N_\mathrm{w}(r_2)$, then the growth factor of the particles of the radius r_1 is equal to r_2/r_1. This method was proposed by A.G. Laktionov[25] for interpretation of the data on the condensation activity measurement by means of the photoelectric counters. For its application two assumptions are to be met:

1. All particles of the same size have close condensation activity (i.e., the growth factor is indeed the function of size).

2. If $r_{1\mathrm{d}} < r_{2\mathrm{d}}$, then $r_{1\mathrm{w}} < r_{2\mathrm{w}}$ (where the index d is related to dry aerosol and w to the wet one).

Practical realization of the Laktionov method becomes more difficult because the particle size distribution is retrieved for the discrete set of radii. So, the interpolation of the inverse function $r_\mathrm{w}(n)$ by cubic splines at the points $N_i = N_\mathrm{d}(r_i)$ was used for determination of the growth factor.

The example of the growth factor as a function of size, obtained in such a way, is shown in Fig. 6. One should note that the estimate of the growth factor of particles with the radius larger than 0.4 μm becomes less reliable due to the prevalent contribution of particles of the radii from 0.1 to 0.3 μm to the measured optical characteristics.

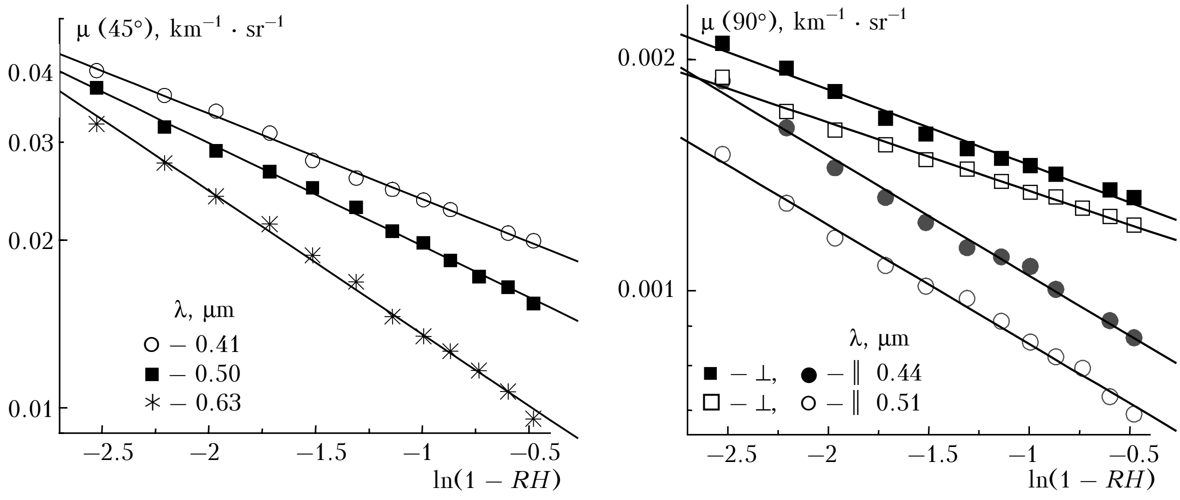


Fig. 4. The measured optical characteristics as functions of the relative humidity of air.

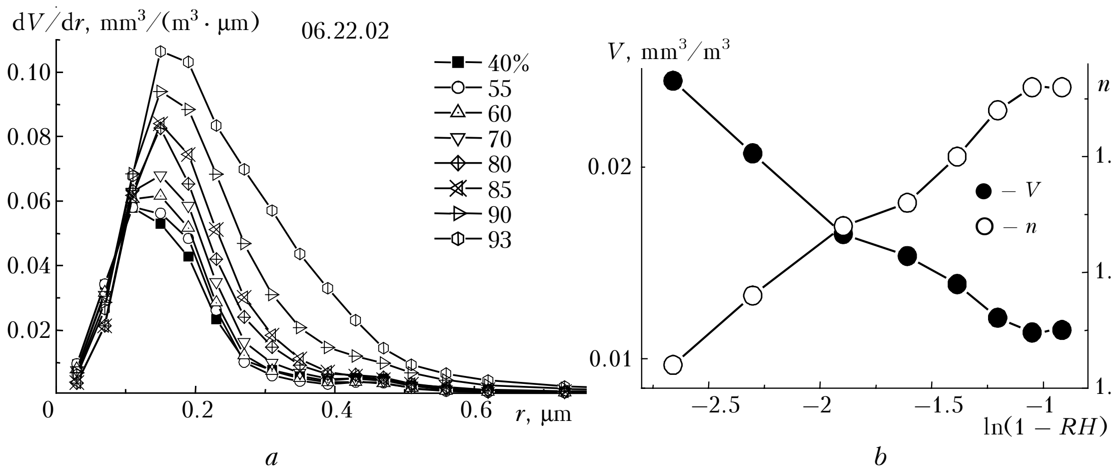


Fig. 5. Transformation of the particle volume size distribution (*a*), volume concentration, and the refractive index (*b*) with the change of relative humidity of air.

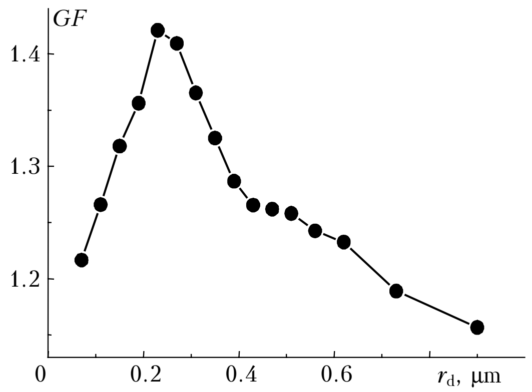


Fig. 6. The growth factor of aerosol particles as a function of the size of dry particle in the range of the air relative humidity from 30 to 80%.

4. Thermooptical investigations of the submicron aerosol

The idea of applying the technique of thermoanalysis, widely used in analytic chemistry for investigation of the properties of aerosol particles, appeared in the 60s and further was developed by N.I. Yudin and Yu.S. Lyubovtseva[26] for optical measurements under atmospheric conditions (thermooptical method). At the same time, measurements of the scattering coefficient[3] or the coefficient of directed scattering at a single wavelength[2,24] allow only to qualitatively estimate the transformation of the total volume of submicron particles during heating.

An example of the change of the optical characteristics at heating is shown in Fig. 7.

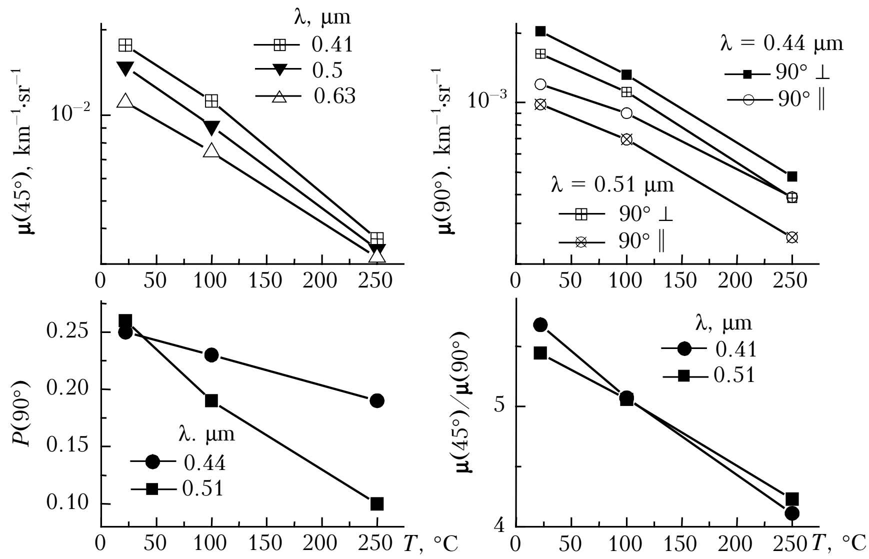


Fig. 7. The measured optical parameters as a function of temperature: μ is the directed scattering coefficient, *P* is the polarization degree.

The particle volume size distributions at different stages of heating are shown in Fig. 8*a*.

Analogously to the growth factor, one can introduce the volatility factor

$$VF = \frac{r(T = 20°C)}{r(T = 100°C)} \tag{4}$$

and to use the same technique for its determination, as for the growth factor. The dependences of the volatility factor on the size of unheated particle, obtained from the solution of the inverse problem, are shown in Fig. 8*b*.

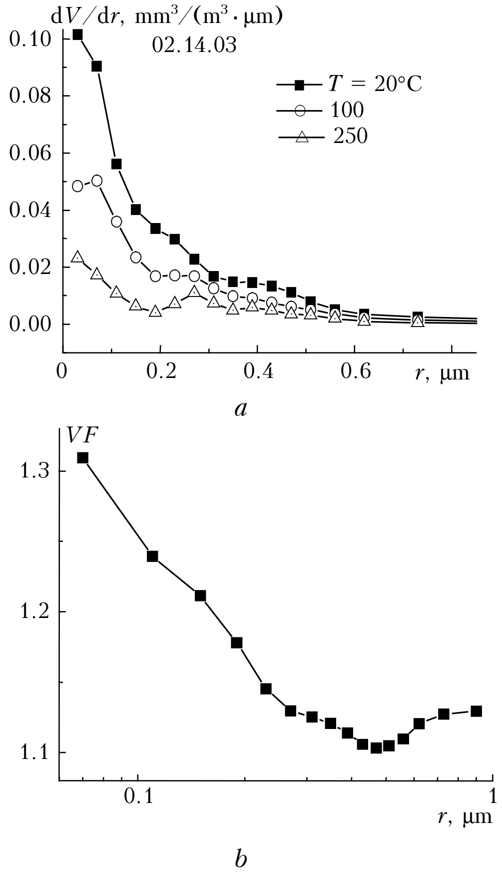


Fig. 8. Transformation of the particle volume size distribution at heating from 20 to 250°C (*a*) and the volatility factor of the aerosol particles in the temperature range from 20 to 100°C (*b*).

On the average, the tendency of decreasing the volatility factor as the size increases is observed in the range from 0.07 to 0.4 μm.

Conclusions

Thus, the method of active nephelometry combining the measurements of the aerosol light-scattering characteristics at artificial influence on its properties with the subsequent solution of the inverse problem makes it possible to essentially extend the capabilities of the experimental studies of the aerosol.

Let us emphasize that, as applied to atmospheric aerosol, the important component of the approach developed is the principle of separate study of the processes of variability of the dry matter of aerosol particles under the effect of the external geophysical factors added by improved investigation of the physicochemical properties of particles by means of the artificial influences.

We see the prospects of the development of this approach in extending the capabilities of the nephelometric setups for investigation of the larger particles. This requires a serious modification of the characteristics of the air sampling paths, the increase of the scattering volumes, expansion of the wavelength range, and the set of the parameters to be measured. Additional prospects of the improved study of the aerosol properties can be realized by use of other methods for controlled influence on particles in the experiments, for example, at investigation of the aerosol condensation activity under the effect of different vapors, electric fields, etc.

Acknowledgments

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