Some statistical regularities in variations of optical and microphysical characteristics of near-ground aerosol

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Results of spectropolarimetric measurements of optical and microphysical characteristics of near-ground aerosol are given. Daily measurements were carried out during February 1999 and February–May 2000 at the Zvenigorod Scientific Station, Institute of Atmospheric Physics, Russian Academy of Sciences. The spectropolarimeter recorded polarization components of light scattered by the near-ground aerosol at three scattering angles φ: 45, 90, and 135° in the wavelength range $\lambda = 0.4–0.76$ μm with spectral resolution $\Delta\lambda \approx 10$ nm. The instrument was equipped with a low-temperature heater for heating the incoming air. The instrument made it possible to estimate the aerosol condensation activity (the Hanel parameter χ) from measured characteristics of wet and dry aerosol. The distribution of χ is found to be close to Gaussian with a variance of 0.3 and mean $\chi = 0.4–0.5$. The correlation between χ and the scattering coefficient is low; however, a spread in values of χ increases markedly with a decrease of the scattering coefficient. The inverse problem was solved for the main body of data obtained for both wet and dry aerosol, i.e., the volume distribution of particles and the complex refractive index of the particulate matter were obtained, which made it possible to estimate the condensation growth factor of particles. Based on the principal-component statistical analysis of the growth factor, a regression relation between the growth factor magnitude and relative air humidity was obtained, which is a microphysical justification of the Hanel formula.

Introduction

Optical and microphysical models of the near-ground aerosol[1–4] constructed in the 70s in IAP RAS and IAO SB RAS are based on the data obtained with the first generation of devices characterized by the so-called open scattering volume, which varied from approximately 10 dm^3 for Stokes-polarimeter of IAP and 100 dm^3 for aureole meters to 1000 m^3 for transparency meters. Practically, the whole spectrum of aerosol particle sizes up to tens of microns participated in formation of working signals. A necessity of carrying out round-the-clock measurements has led to development of the next generation of devices with closed volumes. The payment for size decrease of the polarimeters and nephelometers was a significant decrease (up to several tens of cm^3) of their working volumes. As a consequence, only particles with sizes less than 1.5–2 μm participate in formation of working signals, the passing of larger particles is perceived by the device as a noise splash. This hinders the direct comparison of data obtained with different devices.

The first widespread microphysical model of the near-ground aerosol was the model with the inverse-power particle size distribution. Measurements with photoelectric counters, on the one hand, have confirmed that in the first approximation the model represents principal peculiarities of the particle size spectrum, and, on the other hand, have shown that noticeable deviations from the inverse power law are often observed. They become especially pronounced, when dealing with the volume size distribution. Particle distributions are represented just in the form dV/dr for illustration of the more realistic three-fraction Wittby model, which appeared in the middle of the 70s.

Investigations of smoke aerosol, carried out at the IAP and IAO[5–7,12] smoke chambers, have shown that there are many common features in formation of both smokes and atmospheric submicron aerosol, because two principal mechanisms of particle growth, namely, condensation of aerosol-producing vapors and coagulation, dominate in both cases. The efficiency of the coagulation growth of particles sharply decreases in both cases at $r > \sim 0.2$ μm. In this connection, size distribution functions dV/dr for both natural and smoke aerosols at $r > 0.2$ μm decrease in the first approximation according to the inverse power law. It is quite well fulfilled for natural aerosol in the range of r from 0.2 to 0.8 μm.[9] Therefore, both models, the inverse power law and the lognormal Wittby law provide for similar results in calculations of aerosol optics.

However, it should be noted that the model from Ref. 1 does not quite adequately represent mean features of particle growth as the scattering coefficient σ increases. The model gives a noticeable increase of the modal radius of particles. Analogous microphysical model[9] was constructed by us on the basis of the particle size distribution data obtained from the direct solution of the integral equation. It has shown that the position of the maximum of distribution dV/dr weakly changes as the scattering coefficient increases, and the effective radius of particles increases due to the decrease of the slope of the distribution fragment for $r = 0.2–0.8$ μm which is satisfactorily approximated by the inverse power law.

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When analyzing the efficiency of any model, it is important to outline the class of events, in which it does not work, and to indicate the reasons of this. There are at least two types of atmospheric situation that essentially (up to two times for σ magnitude) deviate from the model. They are: dense finely dispersed hazes formed under conditions of stable anticyclone and the aerosol in the rear part of the cold cyclonic front. For the former, the measured polarization components more or less agree with the model values at approximately halved magnitude of the scattering coefficient. In fact, this means a violation of mean tendency in the change of one of the parameters reconstructed by the microphysical model,[1] i.e., the particle number density, N. The second group of events deviates from the model mainly because of underestimated values of the polarization components due to the effect of non-sphericity of coarse particles, the contribution of which sharply increases in such situations.

The IAP[1] microphysical model of the near-ground aerosol was obtained through inversion by the G.V. Rosenberg method[10] of angular dependences of four components of the scattering phase matrix measured at the wavelength $\lambda = 0.55$ μm The particle size distribution was set by the lognormal function. Analysis of relations between optical characteristics and microphysical parameters retrieved by this technique has shown that the retrieved refractive index n correlates well with the maximal value of the degree of ellipticity[11] of the polarization f_{43}, i.e., the value of n is mainly determined from the maximum of f_{43}.

It was shown[8] that f_{43} for certainly non-spherical particles of the dust haze is much less than the model one. As the input parameter σ of the models decreases, the average relative humidity Rh decreases, and the shape of particles more and more deviates from the sphere and the measured value of f_{34} – from that calculated for spheres. Hence, the values of the refractive index obtained from the inversion are underestimated. It was earlier suggested[13] to use f_{21} (the degree of linear polarization) for estimation of the refractive index. We successfully applied this parameter to determine n of the matter of smoke aerosols.[5,12] An advantage of f_{21} over f_{43} is that the degree of linear polarization is less sensitive to the particle shape. So, the use of two first components of the scattering phase matrix or polarized components of the scattering phase function to solve the inverse problem provides for more realistic estimate of n and seems to be more expedient.

In this paper, the emphasis is on the problem of statistical modeling of such important characteristic of the atmospheric aerosol as its condensation activity. Besides, a possibility of parameterization of the spectral dependences of some near-ground aerosol optical characteristics is analyzed.

Instrumentation

Remind briefly main specifications of the device. The spectral polarimeter records the polarization components of the scattering phase function of the near-ground aerosol at three scattering angles φ: 45, 90, and 135° in the wavelength range $\lambda = 0.4 - 0.76$ μm with spectral resolution $\Delta\lambda \approx 10$ nm. The light source of the device is based on the monochromator of a screen type and provides for quasi-continuous scanning of the wavelength with a controllable step. The working volume of the device at a flow rate of the incoming air of about 1 m/s and the time of signal averaging of about 1 sec is of the order of 1 liter.

A low-temperature heater of the incoming air and the control for its temperature allow one to estimate the aerosol condensation activity. The control for the device and the data collecting are computerized. The spectral polarimeter provides for information sufficient for setting the complex inverse problem – retrieving the particle size distribution function (it was mentioned above that the volume size distribution dV/dr is an optimal representation) and estimating the real and imaginary parts of the complex refractive index of the particulate matter. To solve the inverse problem, a grid of 45 kernel functions was calculated (9 values of the real part of the refractive index in the range $n = 1.35–1.59$ with the step of 0.03 and 5 values of the imaginary part in the range 0–0.02 with the step of 0.005). The radius range was from 0.05 to 1.5 μm. The inverse problem was solved by the iteration method[9] for each kernel function, and the value of the complex refractive index was estimated by the minimum discrepancy method.

Some preliminary notes

1. It is well known that main characteristics of the near-ground aerosol have a well-pronounced seasonal behavior, just which restricts the duration of a measurement cycle to approximately two months. Minimal duration of the cycle is about 1 month.

2. The spectral polarimeter is installed in a laboratory room, so aerosol in the collector can be heated even if the heater is switched off. The magnitude of the heating in a cold season is from 2 to 5°C. So, in winter we deal with fairly dried aerosol.

3. The value of the scattering coefficient σ at $\lambda = 0.55$ μm is commonly used as an input parameter of the aerosol optical models. We consider the value of the directed scattering coefficient $D(\varphi = 45°, \lambda = 0.55$ μm) as an input parameter, using its close correlation with σ.

4. For obviousness, we represent D in units of Rayleigh (molecular) scattering

$$D_{\text{Rel}}(\varphi = 45°, \lambda = 0.55\ \mu\text{m}) = 0.001\ \text{km}^{-1} \cdot \text{sr}^{-1}.$$

5. The device scattering volume is about 1 liter. This reduces the real sensitivity of the spectral polarimeter in statistically mean situations at the rate of air pumping through the chamber ~ 0.5 m/s and the time of digital averaging ~ 1 s to a particle size range 0.05–1.5 μm.

6. A set of 120 values is obtained in each realization during measurements. For convenience of

solving the inverse problem and statistical analysis by the least square method, a smoothed curve (8 points in the range $\lambda = 0.4$–0.75 μm with a step of 0.05 μm) was constructed for each polarized component.

7. There is no doubt today that most measurement errors are due to spatial-temporal instability of the near-ground aerosol. The magnitude of these errors is rare less than 5%, most often it is from 5 to 10%. Spectral dependences both for dry and wet aerosol were recorded in pairs (direct and reverse spectral scans). The records, where the differences in a pair exceeded 15%, were rejected.

Measurement conditions

In 1999, daily measurements started in February and continued till the middle of March. Usual time of measurements was from 9 a.m. to 11 p.m. From 5 to 10 realizations were recorded a day depending on the situation (stable conditions or a change of air mass). Total quantity of spectra exceeded 200. The second experiment under the ARM program started in February 2000 and continued till June 2000. Meteorological conditions in February and March were unusual for that season. Moscow Region turned out to be at the boundary of cold and moderate air masses, and Moscow was found near the centers of a sequence of the cyclones passed. Their atmospheric fronts of occlusion had narrow warm sectors, and there were only a few hours between warm and cold fronts. Relative humidity Rh in the daytime often decreased to 40–50%. The interval between cyclones usually was 2–3 days.

The analysis, carried out by V.N. Sidorov, showed that the characteristic values of aerosol scattering in February and March were approximately a half of the long-term mean values.[14] In April, on the contrary, they were higher than the mean values. Snow cover disappeared in the middle of April. Unusually dry cold weather lasted almost all May. Arctic air was very dry (relative humidity often was less than 30%), the scattering coefficient did not exceed 2–5 Rayleigh scattering units. To estimate properly the aerosol condensation activity using the Hanel formula, it is necessary to have the initial relative humidity at least not less than 50%. The effect of humidity on aerosol at lesser Rh values is often masked by spatial-temporal inhomogeneity of aerosol. In this connection, two data arrays were formed from the data of 2000: February–March (the first) and April (the second). It was impossible to use the data obtained in May for estimating the aerosol condensation activity because of low relative humidity of air.

Discussion of the results

The Angström formula is often used for parameterization of spectral dependences of the scattering coefficient σ and the aerosol optical thickness τ: $\sigma, \tau \sim \lambda^{-\alpha}$. We have analyzed a suitability of the approximation of $D(\varphi = 45°, \lambda)$ separately for the data of February 1999, February–April 2000, and May 2000.

It was found that the main body of realizations of the two first arrays in the wavelength range $\lambda = 0.4$–0.65 μm (more than 90%) is approximated by the Angström formula with the rms error of 2–5%. These errors in May are noticeably greater, i.e., about 10%. Measured values of $D(\varphi = 45°, \lambda)$ at $\lambda > 0.65$ μm exceed the calculated ones, and the difference increases as the wavelength and the scattering coefficient increase. The parameter α (calculated as the angular coefficient of the regression line) varied from 0.5 for dense wet hazes to 2.5 for conditions of high transparency.

Contrary to the expectations, a high correlation between α and $\ln[D(\varphi = 45°, \lambda = 0.55\ \mu m)]$ was noticed, the correlation coefficient R for two first arrays was 0.71 and 0.75, respectively. So high correlation describes a half of the total variance of α and can be used for retrieving α from D: $\alpha = \text{const} - \beta \ln(D)$. The values of β in both arrays are close, 0.45 and 0.43, respectively, the values of the standard deviation $\Delta\alpha = \pm 0.3$.

Correlation between α and $D(\varphi = 45°, \lambda = 0.55\ \mu m)$ in May 2000 was somewhat worse, $R = 0.5$, its "destroyers" were the points corresponding to anomalous days of two first weeks of May (strong north wind up to 10 m/s, the temperature about 0°C, $Rh < 40\%$, dry upper layer of soil, the scattering coefficient of 2–3 Rayleigh scattering units). They have formed a separate area with $\alpha = 1$–1.5 and $D(\varphi = 45°, \lambda = 0.55\ \mu m)$ was 2–5 Rayleigh scattering units. Evidently, the decrease of α was related to the enhanced content (wind emission) of coarse soil particles.

The empirical Hanel formula has been known already for thirty years. It connects the atmospheric aerosol scattering coefficient σ and Rh:

$$\sigma = \sigma_0(1 - Rh)^{-\chi}, \quad (1)$$

where σ_0 is the scattering coefficient of dry particles, χ is the Hanel parameter interpreted as the parameter of aerosol condensation activity. It was shown[15,16] that the formula is also suited for description of variability of the directed scattering coefficient D. If $D_{1,2}$ were measured at $Rh_{1,2}$, respectively, then

$$\chi = \ln(D_1/D_2)/\ln[(1 - Rh_2)/(1 - Rh_1)]. \quad (2)$$

In fact, this is a "short" method for determining χ. Main errors in estimating χ are attributed to inaccuracy in determining the numerator of the formula (2). At low initial $Rh \sim 50\%$ $D_1/D_2 \sim 1.1$, and the spatial-temporal inhomogeneity of aerosol characteristics even under favorable stable conditions is rare less than 2–3%. So, when analyzing the aerosol condensation activity, all realizations with initial values of humidity in the device chamber $< 50\%$ were removed from the consideration.

As for the accuracy of measurements of Rh, let us note the following. Initial values of Rh (for wet

aerosol) were obtained at positive temperatures measured with a psychrometer and at negative temperatures – with the hygrometer calibrated by this psychrometer. Relative humidity in the device chamber was determined by the following scheme: (the air temperature T + relative humidity) *in situ* → the absolute humidity of air + the temperature in the device chamber $T_{1,2}$ → relative humidity in the chamber $Rh_{1,2}$. Since the basis for both values of $Rh_{1,2}$ was the same value of the absolute humidity, and the temperature measurement was quite accurate ($\Delta T = (\pm 0.2)$°C), the error in determining the denominator of the formula was noticeably corrected, at any case, it was much less than it could be at independent determination of $Rh_{1,2}$.

Thus, two statistical ensembles were formed from the winter and spring 2000 measurements: February–March and April. Such a division takes into account the aforementioned essential differences in the measurement conditions. These differences are reflected in statistical characteristics of two ensembles. The preliminary data processing has resulted in an individual array of $\chi(\varphi = 45°, \lambda = 0.54\ \mu m)$ values. Because of importance of this parameter, we analyze its statistical characteristics in more detail. For smoothed spectral dependences $D(\varphi = 45°, \lambda)$ the corresponding dependences $\chi(\varphi = 45°, \lambda)$ were constructed, and statistical analysis of these ensembles was carried out by the principal component method.

The distributions $\chi(\varphi = 45°, \lambda = 0.54\ \mu m)$ for both ensembles are normal (see Fig. 1) with equal variances (0.3) and noticeable different means: $\chi = 0.45$ for the first ensemble and $\chi = 0.35$ for the second. The right wing of the distributions decreases slower than the model one, therefore, the model means are lower than the means over the ensembles: $<\chi> = 0.5$ and 0.4, respectively. There was found a presence of seasonal behavior and a spring maximum,[19] but that was not the only reason of the difference between the means of two ensembles in the considered data array.

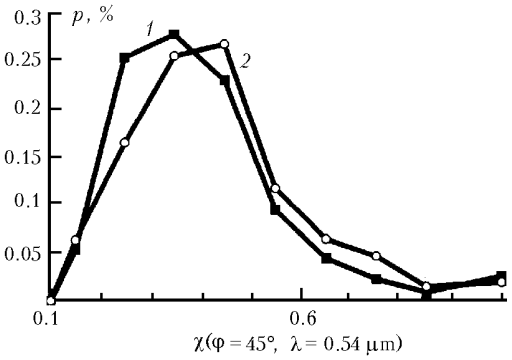

Fig. 1. Histograms of the Hanel parameter $\chi(\varphi = 45°, \lambda = 0.54\ \mu m)$ for the April (*1*) and February–March (*2*) ensembles.

We have analyzed a connection between the parameter of condensation activity and the magnitude of the scattering coefficient. It is found that there is a significant correlation between them ($R > 0.3$). The scatter of points in the two-dimensional diagram $D(\varphi = 45°, \lambda = 0.54\ \mu m) - \chi(\varphi = 45°, \lambda = 0.54\ \mu m)$ has a characteristic shape of right triangle with the legs parallel to the coordinate axis. The scatter of points is insignificant at high values of the scattering coefficient $D > 20$ (there are no values $\chi > 0.5$ and $\chi < 0.3$) and quickly increases as D decreases. The range of variations at $D < 5$ is 0.1–0.8. It is clear that the boundary values, especially the upper one, were obtained with a noticeable error, and the real range is less, but the tendency of the increase of χ averaged over the narrow range of variations of D is observed as the value of the scattering coefficient decreases.

The ensemble-average spectral dependences $\chi(\varphi = 45°, \lambda)$ and two first eigenvectors of covariance matrices of the ensembles are shown in Fig. 2. In both cases the first vector describes about 75% the total variance, and the second describes about 15%. Comparison of the mean values of $\chi(\varphi = 45°, \lambda)$ makes it possible to see both their general features and characteristic differences. Their spectral behaviors at $\lambda < 0.6\ \mu m$ are close, at greater wavelengths the mean for the April ensemble is less. Such behavior of $\chi(\varphi = 45°, \lambda)$ allows one to suppose that the condensation activity of particles quickly decreases as the particle size increases, or that there exists a second fraction in the range of big sizes. With the results of solving the inverse task in hand (retrieved particle size distributions of dry and wet aerosol), we can consider this problem in more detail. Following A.G. Laktionov,[17] we determine the growth factor of aerosol particles as the ratio of radii corresponding to the same value of the integral distribution of "wet" and "dry" aerosol particles. Variations of the particle size distribution density $\zeta(r)$ at variations of relative humidity are equivalent to the change of scale of the radius axis according to the law $r' = h(r)$ at the constant integral number density

$$\eta = \int_a^b \zeta(r)\mathrm{d}r = \eta' = \int_{h(a)}^{h(b)} \zeta(h)\mathrm{d}h. \qquad (3)$$

During condensation transformation, the particles pass from the size range $[a, b]$ to the range $[h(a), h(b)]$. Considering η and η' as functions of the upper limit of integration, one can determine the dependence $h(r)$, defined in Ref. 17 as the growth factor of particles, from the condition $\eta = \eta'$. One can avoid the error related to an uncertainty of the lower limit of integration by integrating $\zeta(r)$ from great radii to less ones; and the sharply increasing η and η' very quickly "forget" the initial conditions. The problem is reduced to determination of the upper limit of the integral $r' = h(r)$ by its value from the condition $\eta = \eta'$. For experimental dependences $\eta(r)$ it inevitably leads to one or another form of differentiation of the experimental curve. In our opinion, it is more reasonable to determine $h(r)$ from the smoothed curves using, for example, polynomial interpolation and the least squares method (LSM). As

the argument and the function in LSM can easily change places (constructing the inverse function), determination of $h(r)$ is not difficult.

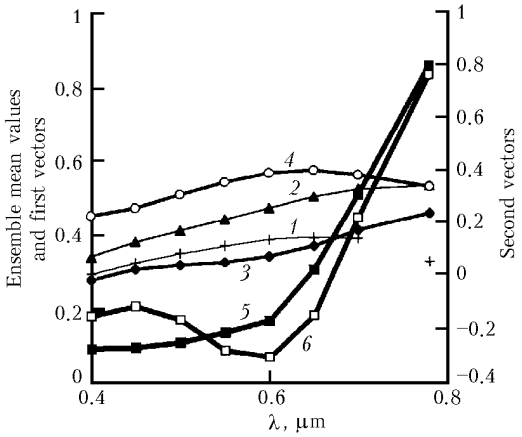

Fig. 2. Principal statistical characteristics of spectral dependences of the Hanel parameter $\chi(\varphi = 45°, \lambda)$ for the February–March and April ensembles. The ensemble average (*2*), the first (*1*) and the second (*5*) eigenvectors for February–March, and *4*, *3*, *6* for April, respectively.

Analysis of the curves $h(r)$ shows that they have a maximum in the range $r = 0.2$–0.5 μm. The ensembles of $h(r)$ formed from the initial data $D(\varphi, \lambda)$ for statistical analysis are adequate to the two ensembles considered above. The means $h(r)$ and two first eigenvectors are shown in Fig. 3. The first vectors of both ensembles describe about 90% the total variance, and the second vectors describe about 5%. The first eigenvector of a single-parameter ensemble follows the behavior of the total variance. The second vectors of the ensembles describe the relatively small quantity of situations when the growth curve has a maximum in the size range $r = 0.2$–0.3 μm. The fact that the obtained statistical mean position of the maximum of the growth curve slightly deviates from the data of Ref. 17 and 18, can have several reasons, for example, the difference in the processes of assimilation and dissimilation of moisture, the difference in techniques, etc. Optical method for estimating χ uses optical manifestations of particle draining, which consist not only in a decrease of particle sizes but also in a change of their structure.

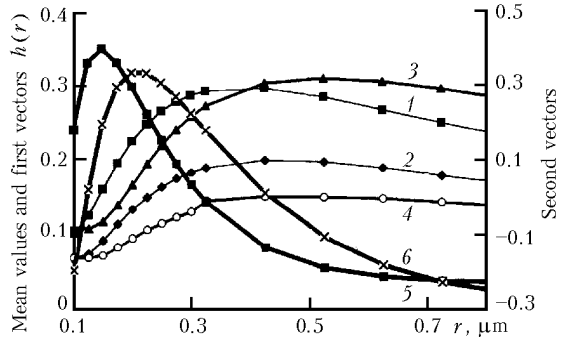

Fig. 3. Main statistical characteristics of the particle growth factor $h(r) = r(Rh)/r_0$ for two ensembles. The ensemble average (*2*), the first (*1*) and the second (*5*) eigenvectors for February–March, and *4*, *3*, *6* for April, respectively.

The final purpose of the analysis by the principal component method is a revealing of connections between the coefficients of expansion of vectors and some easily measured meteorological parameters. This is a usual way of constructing a statistical model. We have analyzed a relation of the first coefficient of expansion of the growth factor $h(r)$ with $dRh = \ln[(1 - Rh_2)/(1 - Rh_1)]$ entering the formula for determination of the condensation activity χ. The correlation diagram (Fig. 4) illustrates this relation. The points from different ensembles are marked by different symbols. The correlation coefficient R for the first ensemble is 0.75, for the second 0.85, and for the total ensemble $R = 0.8$.

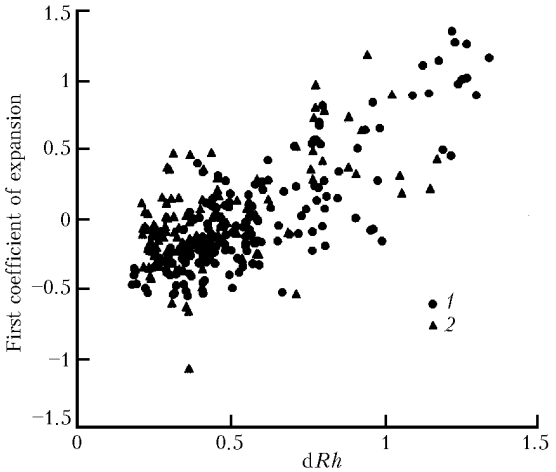

Fig. 4. Correlation diagram of the first expansion coefficient of the particle growth factor $h(r) = r(Rh)/r_0$ and $dRh = \ln[(1 - Rh_2)/(1 - Rh_1)]$ for two ensembles, April (*1*) and February–March (*2*).

Thus, in spite of the differences in the measurement conditions for the ensembles and their statistical characteristics, there is a stable correlation between the considered parameters of both ensembles and dRh, which describes 2/3 the total variance of the first expansion coefficients of the particle growth curves. The point fields of different ensembles almost totally overlap, which is the evidence of universal character of the relation. In fact, the regression in Fig. 4 is the microphysical justification of the Hanel formula.

Conclusions

1. Spectral dependences of the directed scattering coefficient of the near-ground aerosol $D(\varphi = 45°, \lambda)$ in the wavelength range 0.4–0.65 μm are well approximated by the Angström formula. Under standard conditions (except for an enhanced content of dust particles during cold atmospheric fronts) there is a good correlation between the Angström parameter α and the scattering coefficient σ (correlation coefficient $R \approx 0.7$), this relationship can be written as $\alpha = \text{const} - 0.45\ln\sigma$. The standard deviation for α is approximately 0.3.

2. Distribution of the Hanel parameter $\chi(\varphi = 45°, \lambda = 0.54$ μm) is close to the normal law with the

mean of 0.4–0.5 and the variance of 0.3, the mean in February–March is greater than in April. A tendency of increasing range of χ variations as the scattering coefficient σ decreases is revealed.

3. The growth factor of aerosol particles $r(Rh)/r_0$ in the radius range 0.1–1 μm has its maximum in the range r = 0.3–0.4 μm. Statistical ensembles $h(r)$ also are single-parametric, the first vector describes about 90% the total variance, the second one describes the situations when the maximum of $h(r)$ is at $r < 0.3$.

4. Statistical relation of the particle growth factor with the relative humidity of air can be used for estimating $h(r)$. The formula describing this relation is the microphysical justification of the Hanel formula and describes more than 2/3 the total variance of $h(r)$, the residual variance is related to variations in the chemical composition of particles.

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