https://doi.org/10.15407/ujpe66.6.466

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RECOVERY OF PARAMETERS FOR THE MULTIMODAL AEROSOL COMPONENT IN THE ATMOSPHERE FROM SPECTRAL POLARIMETRIC MEASUREMENTS

A method for detecting the major aerosol modes in an atmospheric column and recovering the probable values of the microphysical parameters of their particles from the spectral phase dependences of the sky linear polarization degree has been proposed. A test processing of sky polarization measurements over the location site of the Main Astronomical Observatory of the National Academy of Sciences of Ukraine (Golosiiv, Kyiv) is performed. Two major, coarse and fine, aerosol modes are found in the city atmosphere. The microphysical parameters of those modes are determined assuming the normal-logarithmic distribution function for the particle sizes.

Keywords: atmosphere, degree of linear polarization, aerosol, recovery of parameters.

1. Introduction

Atmospheric aerosol directly affects the physical properties of the atmosphere, thus being an important climatic factor. According to available data [1], aerosol particles smaller than 2.5 μm in size (the $\mathrm{PM}_{2.5}$ standard) are responsible for approximately seven million premature deaths throughout the world every year, with about 400,000 of them in the EU countries. According to moderate estimates for Ukraine made in 2011 [2], the aerosol-induced air pollution is the origin of at least 27 thousand deaths in our country annually, whereas the corresponding GDP losses are estimated to equal 2.6 billion UAH (4% of GDP). At the same time, those values were indicated as, potentially, strongly underestimated because of the lack and low quality of the initial data concerning the air pollution. Therefore, the improvement and development of new methods for monitoring the air pollution with aerosols of various types is a challenging scientific task both in Ukraine and worldwide.

Most often, the atmospheric air quality is monitored at meteorological stations or mobile monitoring laboratories using the contact method, by analyzing the pollution of the near-surface air or atmospheric

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precipitations. This is done, in particular, with the help of optical particle detectors, nephelometers or electrostatic sensors [3]. In this context, promising is the application of passive and active remote methods for the operational monitoring of the atmosphere and atmospheric aerosol in a wide interval of altitudes and over large areas, in particular, on the global scale in the case of satellite-assisted monitoring.

The vast majority of works aimed at determining the altitude distribution and physical parameters of the aerosol component in Earth's atmosphere are performed on the basis of the results of photometric measurements obtained making use of lidars and solar photometers (see, e.g., works [4–9]). Probing the atmosphere only with lidars or measuring the parameters of the luminous flux in narrow spectral intervals with the help of solar photometers allows the results of such measurements to be used for the calculation of the altitude and optical thickness of the aerosol layer, its dynamics and height distribution, but does not allow its optical and microphysical parameters to be determined in more details [10].

For the case of synchronous application of a lidar and a solar photometer that are located at the same observation point, the GARRLiC algorithm for the model analysis of measured data has been developed [11]. As a result, the number of determined characteristics of atmospheric aerosol substantially increased. These are the number of aerosol modes, the type and the parameters of the particle size distribution function, the complex refractive index, the optical aerosol thickness, the single-scattering albedo, the scattering indicatrix (the phase function), the vertical concentration distribution, as well as the inverse scattering and attenuation coefficients.

However, the application of lidars requires that the observation point should be thoroughly equipped, including the installation of a power system for supplying a laser. At the same time, solar photometers, owing to the presence of several spectral measurement channels, need the accurate tracking of the temporal changes in the solar radiation parameters. Therefore, the duration of the monitoring of definite sky areas with their help is restricted, which is associated with the necessity to track the zenith angle of the sun with a given accuracy. Furthermore, the functioning of solar photometers is considerably affected by the spatial and temporal changes in the parameters of separate components in the atmospheric mixture of gases and aerosols, as well as inhomogeneities on the Earth's surface, because of multiple events of light scattering, luminescence, and so forth. A characteristic feature of the indicated devices is their inability to measure the characteristics of low-intensive atmospheric aerosol layers (the "low cloud sky").

At the same time, the analysis of the photo- and polarimetric data obtained for other planets in the solar system is successfully used to determine the parameters of aerosols contained in their upper atmosphere (see, e.g., works [12–18]). Those results testify to a high potential of photo-polarimetric studies and a necessity to develop methods for analyzing the sky polarization in order to determine the physical characteristics of the aerosol component in the Earth's atmosphere [19,20]. Pilot satellite monitoring systems, such as the POLDER device series [21, 22], have demonstrated a high potential of measuring the degree of linear polarization (DLP) of the solar radiation scattered by the Earth's atmosphere in order to determine the properties and the state of atmospheric aerosol on the global scale.

It should be noted that ground-based passive polarimetric measurements gradually began to develop in the last decade. They help to substantially elevate the reliability of determining the aerosol parameters, especially the fine aerosol fraction. However, it is pointed out that the operators of large international observation networks, such as AERONET, still remain reluctant to include polarimetric measurements into the programs of permanent measurements because of their high complexity and a necessity to apply much more efforts to obtain and interpret polarization data [23].

In this work, we proposed a practical method for determining the probable physical parameters for the major modes in the aerosol component of the Earth's atmosphere on the basis of the measurement data obtained for the spectral phase dependences of the DLP of the sky. As an example illustrating the application of the proposed method and the developed kit of special computer program codes, we performed a test analysis of the results of the spectral phase measurements of the DLP of the sky at the zenith above Kyiv, which were reported in work [24].

2. General Definitions and Model Parameters

The degree of linear polarization of the solar radiation scattered by the Earth's atmosphere is determined by the cumulative effect of scattering at gas molecules and aerosol particles [25]. The dependence of the DLP of the sky resulting from a single molecular scattering of light on the Sun position is described by the expression

$$P_R(\alpha) = \sin^2(\alpha)/(1+\cos^2(\alpha)+\delta/(1-\delta)), \qquad (1)$$

where α is the angle between the direction from the Sun to the observed sky area and the direction from this sky area to the observation point (the phase angle), and δ is the depolarization index, which makes allowance for the natural anisotropy of atmospheric gas molecules. Provided that the optical thickness of the atmosphere is small, $\tau_R \ll 1$, the values of $P_R(\alpha)$ are maximum (about 95%) at phase angles of 90° and 270°. For substantially larger τ_R-values, the DLP of the sky gas component decreases owing to the growth of the depolarizing action from multiple scattering [26]. However, the sky measurements at the zenith making no use of light filters demonstrate that, on different days, the values of $P_R(\alpha)$ amount to only 30–70% at the same phase angles [27, 28], which is much less than the DLP value calculated theoretically for a purely gaseous atmosphere. This is a result of the light scattering decrease at aerosol particles, which are always available in the Earth's atmosphere [29].

The DLP of the light scattered by aerosol particles has a complicated phase dependence. It takes both positive and negative values. It depends on the wavelength λ, the nature of the particles, their shape and type, the parameters of the particle size distribution (the radius and the dispersion), and the phase angle (see, e.g., works [30–33]).

The scattering of light at homogeneous spherical particles is rigorously described in the framework of the Mie theory, whereas the problem of finding the phase dependences of the DLP of the light scattered by them can be successfully solved using a computer simulation (see, e.g., work [34]). The research of the characteristics of light scattering by a static ensemble of chaotically oriented homogeneous model particles with the simplest non-spherical shapes (prolate and oblate spheroids, cylinders) showed a significant influence of the particle shape on the parameters of the scattered solar radiation (see, e.g., works [15, 35–37]). However, attempts to simulate the parameters of the light scattered in the Earth's atmosphere by real aerosol particles (salt crystals, snowflakes, soot agglomerates, plant pollen, and others) still face obstacles in the form of the impossibility of the analytical description and the complex character of the algorithmization of such a problem, as well as the extremely high computational difficulties, which require the application of powerful computer clusters.

In the general case, the spectral values of the DLP of the light scattered by the gas-aerosol atmosphere of a planet are presented in the form

$$P(\alpha,\lambda) = \beta(\lambda)P'_R(\alpha) + [1-\beta(\lambda)]\,P_a(\alpha,\lambda,\rho,m), \qquad (2)$$

where α is the phase angle; $P'_R(\alpha)$ the gas component of the DLP calculated taking the influence of the depolarization factor into account [see expression (1)]; $P_a(\alpha,\lambda,\rho,m)$ the aerosol component of the DLP; $\rho = 2\pi a/\lambda$ is the Mie parameter; a the effective radius of aerosol particles; $m = n_r - in_i$ is the complex refractive index (CRI) of the particles; n_r and n are the spectral values of the real and imaginary parts, respectively, of the CRI;

$$\beta(\lambda) = \frac{\sigma_R(\lambda)}{\sigma_R(\lambda)+\sigma_a(\lambda)} \equiv \frac{\tau_R(\lambda)}{\tau_R(\lambda)+\tau_a(\lambda)} \qquad (3)$$

is the relative contribution of the molecular scattering, which depends on the wavelength; and $\sigma_R(\lambda)$, $\tau_R(\lambda)$ and $\sigma_a(\lambda)$, $\tau_a(\lambda)$ are the volume scattering coefficients and the optical thicknesses, respectively, of the gaseous and aerosol components, respectively, in the atmosphere at the wavelength λ [38]. In order to calculate the spectral values of the quantity $\beta_{\text{calc}}(\lambda)$, we took the relevant expression from work [13] and modified it to account for the multimodal character of the aerosol component in the researched atmosphere. As a result, we get

$$\beta_{\text{calc}}(\lambda) = \left[\frac{\sum_{i=1}^{N} k_i\sigma_{ai}(\lambda)}{\sum_{i=1}^{N} k_i\sigma_{ai}(\lambda_0)}\left(\frac{\lambda}{\lambda_0}\right)^4 \times \right.$$
$$\left. \times \left(\frac{1-\beta(\lambda_0)}{\beta(\lambda_0)}\right) + 1\right]^{-1}, \qquad (4)$$

where $\sigma_{ai}(\lambda)$ is the volume scattering coefficient at the wavelength λ for the i-th mode in the aerosol mixture, k_i the weight coefficient for the i-th mode, N the number of aerosol modes, and $\sigma_{ai}(\lambda_0)$ and $\beta(\lambda_0)$ are the values of the corresponding quantities at the wavelength λ_0.

The aerosol component $P_a(\alpha,\lambda,\rho,m)$ of the DLP of the sky includes a component formed by the single scattering and a component formed by the multiple scattering. The latter is insignificant in the long-wave interval of the light spectrum, but it substantially increases in the short-wave interval [39].

3. Parameter Recovery Procedure for the Multimodal Aerosol Component of the Atmosphere

Before describing the algorithm of the proposed method, let us bring together the accepted model restrictions and assumptions, as well as their origins.

a) The results of works [40–44] testify that the normal-logarithmic distribution function for the particle sizes is the most adequate for the majority of the aerosol particle distributions that were analyzed in the Earth's atmosphere. Therefore, just this particle size distribution function was applied in this work. For physical reasons, the size dispersion of particles was considered to be within the interval $\sigma^2 = 0.01 \div 1.0$. We should emphasize however that the choice of the size distribution function for aerosol particles and the indicated interval of σ^2-values are not a matter of principle for the proposed method aimed at determining the aerosol characteristics.

b) For the overwhelming majority of terrestrial aerosols, the imaginary part of the CRI for their particles is small ($n_i < 10^{-3}$) in the visible spectral interval of the solar radiation [44, 45]. Therefore, the second Stokes parameter for the light scattered in the atmosphere is mainly determined by the real part of the CRI of aerosol particles only, which possesses weak dispersion properties [46]. As a result, when modeling the parameters of atmospheric aerosols, we neglected the light absorption by their particles ($n_i = 0$) and did not take the dispersion of the parameter n_r in the studied spectral interval into account. Note that this approximation is not crucial for the proposed method.

c) In spite of the presence of aerosols of various types and origins in the Earth's atmosphere and neglecting the above-mentioned influence of the shape of scattering particles on the scattered light parameters, we, nevertheless, used the model of homogeneous spherical aerosol particles. This choice was done because of the following main reasons.

- The phase dependences of the DLP of chaotically oriented nonspherical particles still reproduce, in general, the corresponding dependences for spherical particles [38, 47].
- In view of the absence of information on the nature and shape of aerosol particles that are available in the examined atmosphere region, the choice of a specific shape and a specific internal structure of model particles is unjustified, being absolutely identical to any other choice. In order to make the analysis correctly, it is necessary, when performing the calculations, to additionally introduce and to consider the functions for the altitude distributions of aerosol particles over their form and structure. However, this task is almost unrealizable in the case of remote measurements.
- It is evident that the experimental dependences of the DLP of the sky are formed due to the light scattering along the line of observation in the whole atmospheric column. Therefore, the choice of the same non-spherical shape for all aerosol particles will be erroneous because of different physical conditions at different altitudes. Moreover, there is a high probability that the nature of aerosols is different at different altitudes.
- Numerous practical measurements of the Earth's atmosphere composition prove that spherical particles prevail in tropospheric aerosols [44]. Also, according to the results of work [48], fine spherical particles dominate in the cloudless urban atmosphere, whereas large spherical agglomerates of particles predominate in the turbid (smoky) environment.
- Chaotically oriented aerosol particles with arbitrary shape and structure, even if those parameters are definitely known, are not static but permanently move in the atmosphere. Under the action of gravity, wind, thermal convection, mutual collisions, and so forth, they permanently change their positions and orientations in space. Therefore, as the measurement time becomes longer, the effective cross-section for aerosol particles with an arbitrary shape gradually approaches the cross-section corresponding to the spherical shape. As a result, the scattering properties of the researched volume in the atmospheric medium also approach the properties of an ensemble of quasispherical particles with the corresponding effective radius and the averaged complex refractive index of a quasihomogeneous substance.
- Elastic scattering of light at homogeneous spherical particles has a rigid theoretical solution in the framework of the Mie theory. This circumstance makes it possible to realize an algorithm for an accurate analysis of initial data and to calculate the

aerosol parameters for a model gas-aerosol medium reducing the number of preliminary assumptions about its real physical properties to a minimum.

• The model of spherical particles is the simplest for computations, which is an important factor when carrying out a mobile ground-based or satellite polarimetric experiment because, ideally, such calculations should be executed in the real-time mode.

d) Owing to numerous remote and contact studies of the properties of the most common aerosol particles in the Earth's atmosphere, the intervals of values for their parameters are known quite well today (see, e.g., work [46]). Our selection of parameter values for the model aerosol was done just within those intervals. However, we should emphasize that, if necessary, those intervals can undoubtedly be extended without putting any restrictions on our method.

Hence, with regard for the indicated assumptions and restrictions, we adopt the normal-logarithmic distribution function for the sizes of aerosol particles and, in what follows, we are going to determine the dependence of the degree of linear polarization P_a for the light scattered by the model aerosol on the following parameters: the phase angle α, the wavelength λ, the dimensionless Mie parameter ρ, the real part of the complex refractive index n_r, and the particle size dispersion σ^2.

3.1. First stage

While analyzing the data on the DLP of the sky, at the first stage, we apply the model, where the atmosphere is considered as a homogeneous gas-aerosol medium containing only one aerosol fraction, mode 1.

Step 1. The dependences of the DLP of the light scattered by the components of the model medium – the dependence $P'_R(\alpha)$ for the gaseous mode and the dependence $P_{a1}(\alpha, \lambda_0, \rho_1, n_{r1})$ for the polydisperse aerosol mode 1 – are calculated. The values of the parameters n_{r1}, ρ_1, and σ_1^2 are selected consecutively within the corresponding intervals of physically allowable values. The model phase angles α correspond to the experimental measurement angles of the DLP of the sky at the wavelength λ_0 in the long-wave section of the spectral interval, $P_{\text{meas}}(\alpha, \lambda_0)$. Such a choice of the wavelength is associated with a requirement to eliminate the contribution of the multiple scattering to the total value of the DLP of the sky. Then expression (2) is used to determine the dependence $P(\alpha, \lambda_0)$ for the DLP of the light scattered by the model gas-aerosol medium. This dependence is compared with the measurement data for the DLP of the sky, $P_{\text{meas}}(\alpha, \lambda_0)$. The quality of the coincidence is estimated by calculating the sum of squared deviations (SSD) of the model DLP values from the experimental dependence over the whole experimental interval of phase angles (see, e.g., work [49]).

After the parameters of the aerosol mode 1 have been determined, the spectral value of its volume scattering coefficient $\sigma_{a1}(\lambda_0)$ is calculated and fixed. The initial value of $\beta(\lambda_0)$ is found from the tabular data for the spectral values of the gaseous and aerosol components of the optical thickness of the Earth's atmosphere making use of formula (3). Then, after the parameters of the aerosol mode 1 have been determined, this value is corrected. A substantial difference between the initial and corrected values of the quantity $\beta(\lambda_0)$ can be considered as the first indication of the presence of an additional aerosol mode in the analyzed atmosphere. The neglecting of the DLP of this mode brings about an overestimation of the gas component contribution to the DLP of the model medium.

Step 2. At this step, the quality of coincidence of the phase dependences for the DLP of the model gas-aerosol medium and the DLP of the sky is tested at the wavelength λ_1 in the short-wave section of the measured spectral interval. For this purpose, the values of the parameters of aerosol mode 1 determined at step 1 are used. The dependences of the DLP of the aerosol component $P_{a1}(\alpha, \lambda_1)$ and its volume scattering coefficient $\sigma_{a1}(\lambda_1)$ are calculated. After the corresponding phase dependence of the DLP of the gaseous component, $P'_R(\alpha)$, has been determined, expression (2) is used to calculate the dependence of the DLP for the model medium, $P(\alpha, \lambda_1)$, which is compared with the measurement data, $P_{\text{meas}}(\alpha, \lambda_1)$. The value of $\beta(\lambda_1)$ is found from expression (4) with $N = 1$. Then it is corrected until the best coincidence of the phase dependences $P(\alpha, \lambda_1)$ and $P_{\text{meas}}(\alpha, \lambda_1)$ is reached. A substantial difference (by 30% or more) between the values of those dependences at all phase angles is the second indication of the presence of an additional aerosol mode in the studied atmosphere.

3.2. Second stage

The second stage of the analysis is executed provided that the indications of the presence of an additional aerosol mode in the studied atmosphere have been obtained in step 1 and step 2. In this case, the model of gas-aerosol medium is supplemented with fine aerosol mode 2. The polarization properties of the latter have to bring the parameter $\beta(\lambda)$ closer to the physically correct value and provide a better coincidence between the calculated and measured phase dependences of the DLP within the entire spectral interval of measurements.

Step 3. The parameters of the particles in aerosol mode 2 are also first selected at the wavelength λ_0 in the long-wave section of the measurement data. The calculation of the model phase dependence $P_a(\alpha, \lambda_0)$ for the DLP of the aerosol component of the model medium, which consists of two modes, is performed similarly to step 1. But now, this quantity is given by the expression

$$P_a(\alpha, \lambda_0) = k_1 P_{a1}(\alpha, \lambda_0, \rho_1, n_{r1}) + \\ + (1 - k_1) P_{a2}(\alpha, \lambda_0, \rho_2, n_{r2}), \tag{5}$$

where $P_{a1}(\alpha, \lambda_0, \rho_1, n_{r1})$ is the phase dependence for the DLP of the light scattered by aerosol mode 1, $P_{a2}(\alpha, \lambda_0, \rho_2, n_{r2})$ is the same dependence for mode 2, and k_1 is the weight coefficient for mode 1. The parameters of mode 2 are determined following an algorithm similar to that used when selecting the parameters of mode 1 in step 1. The values of the quantities k_1 and $\beta(\lambda_0)$ are corrected after the parameters of mode 2 have been selected. The spectral value of the volume scattering coefficient for mode 2, $\sigma_{a2}(\lambda_0)$, is also calculated.

Step 4. Similarly to step 2, the degree of coincidence between the DLP dependences $P(\alpha, \lambda_i)$ for the model gas-aerosol mixture and $P_{\text{meas}}(\alpha, \lambda_1)$ for the measurement data is tested at the wavelength λ_1 in the short-wave section of the measurement spectral interval. For this purpose, the phase dependences $P'_R(\alpha)$, $P_a(\alpha, \lambda_1)$, and $P(\alpha, \lambda_1)$ are calculated at the wavelength λ_1 using the determined values of the parameters for both aerosol modes, 1 and 2. The initial value of $\beta(\lambda_1)$ is determined according to expression (4) with $N = 2$ and using the calculated spectral values $\sigma_{a2}(\lambda_1)$ and $\sigma_{a2}(\lambda_0)$. As was indicated above, the influence of the multiple scattering on the DLP of the gas-aerosol medium increases substantially in the short-wave section of the spectrum. Neglecting this factor can considerably reduce the degree of coincidence between the calculated, $P(\alpha, \lambda_1)$, and, measured, $P_{\text{meas}}(\alpha, \lambda_1)$, dependences. In this case, the value of $\beta(\lambda_1)$ should be corrected similarly to step 1.

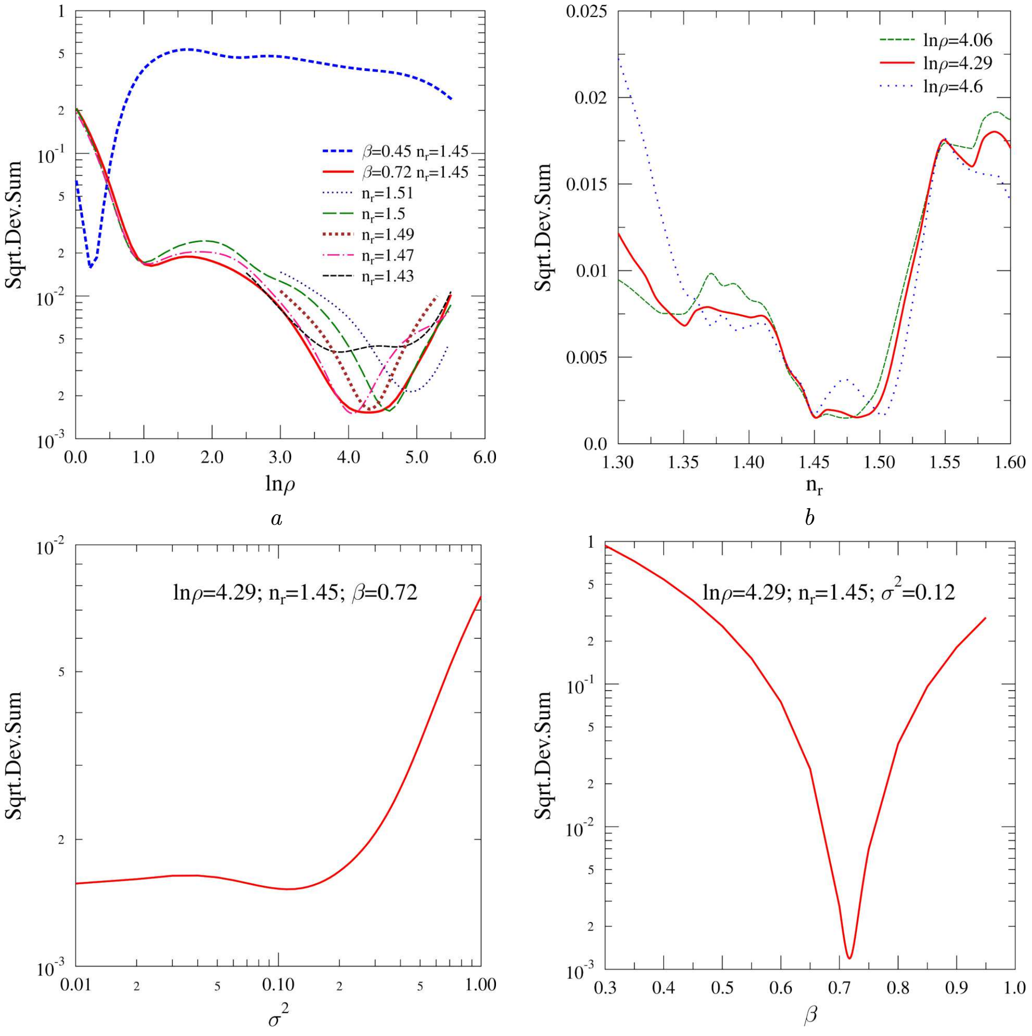
4. Test Analysis of the Measurement Data for the Degree of Linear Polarization of the Sky. Discussion of the Results

Monomode polydisperse aerosol fractions are observed in the Earth's atmosphere very rarely, in particular, above the ocean surface far from the land. The air above the towns almost always contains multimode aerosol compositions of both the natural and anthropogenic origins [44]. Therefore, the expected result of the analysis of the DLP measurements of the sky over Kyiv consisted in the detection of several modes in the aerosol component of the atmosphere.

After having studied the experimental spectral phase dependences of the DLP of the sky presented in work [24] and having analyzed the tabular data of the DLP calculations for the light scattered by a model polydisperse system of homogeneous spherical particles [34], we arrived at the following conclusions.

- The measured phase dependences of the DLP of the sky over Kyiv have substantial spectral differences.
- The form of those dependences testifies to the presence of an aerosol mode, the sizes of particles in which are much larger than the light wavelengths in the measurement spectral interval.
- An appreciable reduction in the values of the DLP of the sky in the short-wave spectral interval of the measurements cannot be a result of only the dispersion properties of the coarse aerosol mode, but testifies to a probable presence of a fine aerosol mode in the atmosphere.
- In order to correctly determine the parameters of aerosol particles in the researched atmosphere, the data of DLP measurements in wide intervals of phase angles and wavelengths are required [38, 50, 51].

Concerning the last item, the following remark should be made. In the analyzed work [24], the spectral measurement data are presented in a rather wide interval of the Sun zenith angles for two observation dates only, April 18 and 26, 1962. In this paper, we present the results of parameters recovery for aerosol particles on the basis of observation data obtained on April 18. Note that, except for the observation data on April 26, the data presented in



a *b*

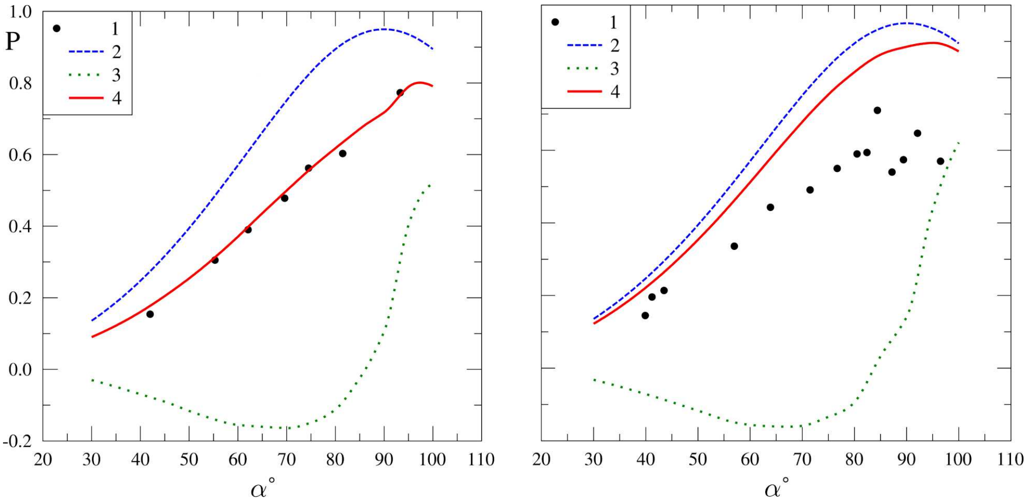
c *d*

Fig. 1. Dependences of the sum of squared deviations (SSD) of the calculated (model) values of the DLP from the experimental ones on the parameters of aerosol mode 1: $\ln(\rho)$ (*a*), n_r (*b*), σ^2 (*c*), and β (*d*)

work [24] for other dates can be used to analyze the DLP dependences only within the phase angle interval from 85° to 96°, which characterizes the light scattering in the upper troposphere and perhaps the lower stratosphere.

At the first stage of our analysis, according to step 1, we determined the parameters for the coarse mode 1 of the atmospheric aerosol in the long-wave section of the measured spectral interval. Figure 1 demonstrates the SSD dependences of the calculated (model) and measured DLP values at the wavelength $\lambda_0 = 578$ nm on the parameters of aerosol mode 1. Note that the smaller SSD value corresponds to a better coincidence between the values of the compared phase dependences of the DLP.

As one can see from Fig. 1, *a*, the best coincidence of the calculated and measured dependences of the DLP of the sky on the parameter $\ln(\rho_1)$ is observed within the interval $\ln(\rho_1) \approx 4.0\div4.6$ for the parameter values $n_{r1} \approx 1.45\div1.50$. The interval indicated for the quantity $\ln(\rho_1)$ is completely overlapped by the SSD dependence calculated for $n_{r1} = 1.45$. Model calculations of the SSD dependence on the parameter n_{r1} carried out for several values of the quantity $\ln(\rho_1)$ taken from the indicated interval showed that the most complete overlap of the



a

b

Fig. 2. Phase dependences of the DLP of scattered light, P, with the wavelengths λ = 578 (*a*) and 390 nm (b): experimental data (*1*), gaseous component (*2*), single-mode aerosol component (mode 1) (*3*), and model medium (*4*)

interval $n_{r1} \approx 1.45\div1.49$ takes place at $\ln(\rho_1) = 4.29$ (see Fig. 1, *b*). Figures 1, *c* and *d* illustrate the graphical dependences of the SSD on the particle size dispersion σ_1^2 and the quantity β, respectively, for the fixed parameter values $n_{r1} = 1.45$ and $\ln(\rho_1) = 4.29$.

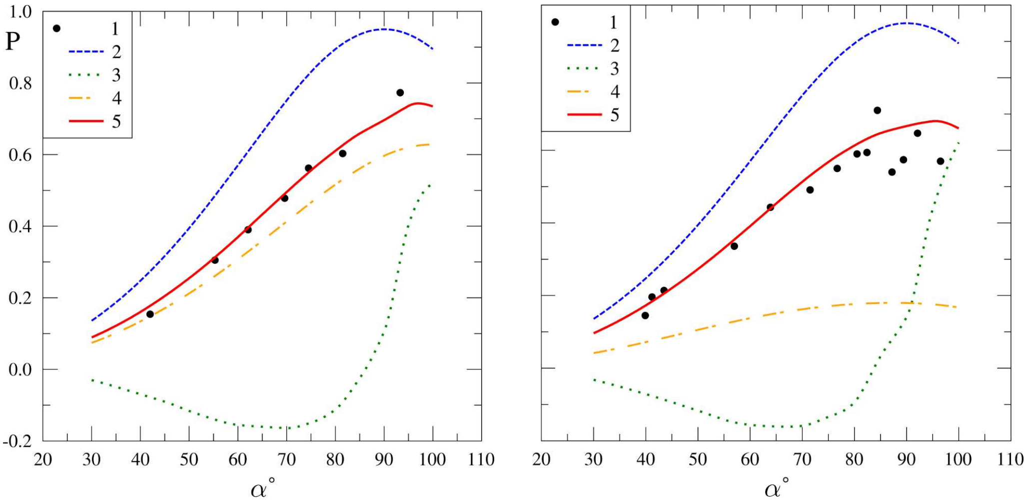
Hence, the most probable values of the parameters for aerosol mode 1 are as follows: the real part of the CRI of the particles $n_{r1} = 1.45 + 0.02/-0.01$, the mean geometric radius of the particles $r_{01} = 6.7 + 2.4/-1.4$ μm, and the particle size dispersion $\sigma_1^2 = 0.12 + 0.01/-0.02$.

In Fig. 2, *a*, the calculated phase dependences of the DLP of the light scattered by the gaseous and single-mode aerosol components and by the model medium, as well as the experimental dependence of the DLP of the sky at the wavelength $\lambda = 578$ nm above Kyiv on April 18, 1962 [24], are depicted. The corrected value $\beta_{\text{spec}}(578\text{ nm}) \approx 0.72$ turned out much larger than the initial value $\beta_{\text{calc}}(578\text{ nm}) \approx 0.5$ (see Fig. 1, *a*), which was calculated on the basis of tabular data taken from work [52] for the spectral values of the gaseous and aerosol optical thicknesses of the Earth's atmosphere at the wavelength $\lambda \approx 578$ nm. This difference is the first indication of the presence of the additional fine aerosol mode 2 in the researched atmosphere.

Afterward, in accordance with step 2, we checked the correspondence of the phase dependence of the DLP for the model medium and the measured data for the DLP of the sky in the short-wave section of the experimental spectral interval. The phase dependence of the DLP at the wavelength $\lambda_1 = 390$ nm calculated for the model medium with aerosol mode 1 turned out shifted upward rather strongly with respect to the corresponding experimental dependence (see Fig. 2, *b*). This result is the second indication of the presence, in large quantities, of the particles of the fine aerosol mode in the researched atmosphere.

At the second stage of our analysis, we added the fine aerosol mode 2 to the model gas-aerosol medium according to the algorithm described in step 3. The selection of parameters for mode 2 allowed us to obtain the physically correct value $\beta(578\text{ nm}) = 0.45$ and substantially approach the phase dependences of the DLP for the model medium to the sky measurement data in the whole spectral interval of experimental data. The corrected value of the weight coefficient for mode 1 was found at this stage to equal $k_1 = 0.22$. Note that, unlike the coarse fraction, the parameters of the fine aerosol mode are determined unambiguously from the results of polarimetric measurements.

The recovered values of the parameters for aerosol mode 2 turned out as follows: $n_{r2} = 1.45 \pm 0.01$, $r_{02} = 0.11 \pm 0.05$ μm, and $\sigma_2^2 = 0.1 \pm 0.05$. The experimental phase dependences of the DLP of the sky at the wavelength $\lambda_0 = 578$ nm and calculated counterparts for the model medium with the bi-



a *b*

Fig. 3. The same as in Fig. 2, but for the two-mode aerosol component: experimental data (*1*), gaseous component (*2*), aerosol mode 1 (*3*), aerosol mode 2 (*4*), and model medium (*5*)

Parameter values for aerosol modes in the sky over Kyiv

Source	Aerosol mode 1		Aerosol mode 2	
	Real part of the refractive index, n_r	Mean geometric radius, r_0, μm	Real part of the refractive index, n_r	Mean geometric radius, r_0, μm
This work	$1.45+0.02/-0.01$	$6.7+2.4/-1.4$	1.45 ± 0.01	0.11 ± 0.005
[55]	1.45 ± 0.05	$6.0+2.0/-4.0$	1.45 ± 0.05	0.2 ± 0.1

modal aerosol component are shown in Fig. 3, *a*. Analogous dependences calculated for the wavelength $\lambda_1 = 390$ nm in accordance with the algorithm described in step 4 are shown in Fig. 3, *b*. The corrected value $\beta_{\rm spec}(390\ {\rm nm}) \approx 0.64$ involves the depolarizing effect of multiple light scattering on the DLP of the sky in the short-wave section of the measured spectral interval. The indicated value is approximately 20% lower than the value $\beta_{\rm calc}(390\ {\rm nm}) = 0.79$ calculated with the help of expression (4) following the algorithm described in step 4.

A considerable scatter of the points in the experimental dependence of the DLP at large phase angles at the wavelength $\lambda = 390$ nm can be associated with the multiple scattering arising due to an almost five-fold increase of the gas optical thickness in comparison with that at the wavelength $\lambda = 578$ nm (see, e.g., work [53]). Furthermore, owing to the growth of the geometric length of the light path, the short-wave fraction of light becomes absorbed (the so-called spectral "reddening") and the signal-to-noise ratio decreases. As a result, the measurement error increases.

Note that the multiple scattering can be taken into account while calculating the DLP of the aerosol component by using the method described in works [13, 54]. However, in this case, the data of synchronous measurements of the photo- and polarimetric characteristics of the sky are required, whereas no photometric measurements were performed in the analyzed work [24].

Table contains the parameter values for aerosol modes 1 and 2, as well as the correspondent atmospheric aerosol characteristics obtained by analyzing the results of synchronous lidar probing and solar photometry measurements obtained in work [55]. One can see that the parameters of two main modes of the aerosol component, which were determined on the basis of the spectral phase measurements of the DLP of the sky over the Main Astronomical Observatory of the National Academy of Sciences of

Ukraine (Golosiiv, Kyiv) performed in April 1962, are close to the aerosol parameters determined from the photometric measurements made at the same site in September 2015. Note also that the weight ratio between modes 1 and 2 revealed in the atmospheric aerosol composition approximately amounts to 1 : 4. This value is close to the results of work [55].

The coincidence of the results obtained by means of essentially different methods and for the data of measurements that are widely spaced in time testifies to a high probability of the reliable recovery of the parameter values for the atmospheric aerosol. In addition, this constancy may point to the stability of the main fractions in the aerosol composition in the atmosphere above Kyiv for a long time interval. However, this assumption requires a statistical confirmation through further measurements of the DLP of the sky and their analysis.

In a number of works devoted to the study of the characteristics of aerosol fractions in the atmosphere over towns (the so-called "urban aerosol") with the help of photometry methods, the characteristic presence of two main, coarse and fine, aerosol modes was also revealed (see, e.g., works [4, 7, 48]). The parameters of the fine aerosol mode were found to be relatively stable in time, with only the volume density of this fraction changing. Note also that the parameters of the fine aerosol mode over other towns are close to the values determined for the atmosphere over Kyiv. On the other hand, the coarse aerosol modes in the atmospheric masses over various towns differ substantially not only in their parameters but often in their nature. Moreover, their characteristics can change considerably and rapidly in time (see, e.g., work [55]).

5. The Kit of Computer Program Codes

In order to simulate the spectral phase dependences of the DLP of sunlight scattered in a gas-aerosol medium, we developed a kit of special computer program codes. The algorithm used to calculate the degree of linear polarization, the scattering coefficient, and the phase function of a polydisperse ensemble of homogeneous spherical particles characterized by a normal-logarithmic size distribution function was developed on the basis of works [34, 56–58]. When verifying the developed kit of programs, the correctness and the calculation accuracy of the angular functions and the Mie amplitude coefficients, as well as the values of the phase function, the DLP, and the volume scattering coefficient, were compared with the calculation results obtained in the works cited above. The software package provides a calculation accuracy of 10^{-8}.

6. Conclusions

An efficient practical method has been proposed to recover the parameter values for the multimodal atmospheric aerosol component on the basis of the results of spectral phase measurements for the degree of linear polarization of the sky. The characteristics of the aerosol component in the sky over Kyiv, which were recovered by us in the course of verification of the method, were found to have a high degree of coincidence with the results of interpretation of lidar observations synchronized with solar photometry measurements. An assumption was made about the possible presence of two main modes in the aerosol component in the atmosphere over Kyiv for a long time interval.

The method proposed for the determination of the parameters of the aerosol component in the Earth's atmosphere is optimal for the analysis of the data obtained via the ground measurements of the DLP of the cloudless sky at the zenith and in the Sun's almucantar. This method may also be adapted to determine the parameters of stratospheric aerosol when processing the measurement data on the DLP of the atmosphere within a wide interval of phase angles obtained from artificial Earth satellites. In this case, the measurements must be performed in the UV range of light, where the light absorption by the ozone layer reliably cuts off the effect of light scattering at the unpredictably and rapidly changing Earth's surface and tropospheric clouds.

The authors express their sincere gratitude to Dr. Sci. in physics and mathematics O.V. Morozhenko for valuable advices and critical remarks during the discussion of the proposed method and the interpretation of the obtained results.

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Received 20.06.20.
Translated from Ukrainian by O.I. Voitenko

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МЕТОД ВІДНОВЛЕННЯ ПАРАМЕТРІВ БАГАТОМОДОВОЇ АЕРОЗОЛЬНОЇ СКЛАДОВОЇ В АТМОСФЕРІ ЗА ДАНИМИ СПЕКТРАЛЬНИХ ПОЛЯРИМЕТРИЧНИХ ВИМІРІВ

Запропоновано метод виявлення основних мод аерозолю в атмосферному стовпі й відновлення ймовірних мікрофізичних параметрів його частинок за даними вимірювань спектральних фазових залежностей ступеня лінійної поляризації неба. Виконана тестова обробка даних вимірювань поляризації неба над позицією ГАО (Київ, Голосіїв, Україна). У міській атмосфері виявлено дві основні аерозольні моди: грубодисперсну й дрібнодисперсну. Відновлено мікрофізичні параметри цих мод для нормально-логарифмічної функції розподілу частинок за розмірами. У грубодисперсної моди дійсна частина показника заломлення $n_r = 1{,}45 + 0{,}02/-0{,}01$, середньо-геометричний радіус частинок $r_0 = 6{,}7 + 2{,}4/-1{,}4$ мкм, дисперсія $\sigma^2 = 0{,}12 + 0{,}01/-0{,}02$, ваговий коефіцієнт цієї моди в аерозольній суміші $k_1 = 0{,}22$. У дрібнодисперсної моди $n_r = 1{,}45 \pm 0{,}01$, $r_0 = 0{,}11 \pm 0{,}005$ мкм і $\sigma^2 = 0{,}1 \pm 0{,}05$. Спектральні величини відносного внеску газового розсіяння (на дату спостережень): $\beta(578\ \text{нм}) = 0{,}45$ і $\beta(390\ \text{нм}) = 0{,}64$.

Ключові слова: атмосфера, ступінь лінійної поляризації, аерозоль, відновлення параметрів.