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O.S. OVSAK, V.M. VASHCHENKO, A.P. VID'MACHENKO 1,3

- ¹ Main Astronomical Observatory, Nat. Acad. of Sci. of Ukraine (27, Akademika Zabolotnogo Str., Kyiv 03143, Ukraine; e-mail: ovsak@mao.kiev.ua)
- ² Interdepartmental Center for Fundamental Research in Energy and Ecology (1, Shevchenka Ave., Odesa 65044, Ukraine; e-mail: nucleoroid@gmail.com)
- ³ National University of Life Resources and Environmental Management of Ukraine (15, Heroiv Oborony Str., Kyiv 03041, Ukraine; e-mail: vida@mao.kiev.ua)

RECOVERY OF THE PARAMETERS OF THE ATMOSPHERIC GAS-AEROSOL ENVIRONMENT ABOVE THE POSITION OF CESAR OBSERVATORY, THE NETHERLANDS

With the method previously proposed by the authors for recovering the parameters of the multimode aerosol component in the atmosphere, the data of polarization measurements of the sky above the position of CESAR Observatory, the Netherlands, at 14:55 UTC on July 9, 2013, have been analyzed. The presence of two aerosol modes with the normal-logarithmic particle size distribution is revealed, and some of their microphysical parameters are recovered. The following parameter values are determined for the coarse-dispersed mode: the real part of the refractive index $n_r = 1.53$, the effective radius of the particles $r = 0.9 \ \mu m$, the dispersion $\sigma^2 = 0.36$, and the weighting factor of this mode in the total degree of linear polarization of the aerosol mixture $coef_1=0.1$. For the fine-dispersed mode, $n_r=1.49,\ r=0.11\ \mu m$, and $\sigma^2 = 0.4$. The quantitative ratio between the indicated aerosol modes in the air above the observation position is estimated to be approximately 1:9. The spectral values of the relative contribution of gas scattering were determined: $\beta(870 \text{ nm}) = 0.3 \text{ and } \beta(675 \text{ nm}) = 0.46$. The incorrectness of using the single Rayleigh scattering model in the short-wavelength region of the visible light spectrum at high air saturation levels with aerosol has been pointed out. A substantial influence of multiple light scattering on the results of polarimetric measurements at a wavelength of 441 nm under the specified atmospheric conditions of sky observation has $been\ demonstrated.$

Keywords: earth's atmosphere, polarization measurements, degree of linear polarisation, aerosol, recovery of microphysical parameters.

1. Introduction

A significant impact of human activity on the chemical composition and aerosol saturation of the at-

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mospheric layers above the cities and territories with developed industry, powerful mining enterprises, large transport interchanges, highways, and, so forth, requires the implementation of routine procedures for environmental air monitoring. Furthermore, manmade and natural disasters happen rather frequently and substantially expand the indicated activity field for relevant municipal departments and units of state emergency services. Complementing contact methods applied to analyze gas and aerosol components in the atmosphere, methods for remote study of Earth's at-

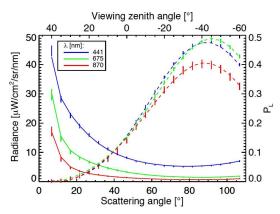


Fig. 1. Measurement results (vertical error bars) and the best fittings of the spectral brightness Radiance (solid curves) and the linear polarization degree P_L (dashed curves) as functions of the scattering angle in the main plane. CESAR Observatory position at 14:55 UTC on July 9, 2013 [7]

mosphere have been developing in recent decades at an extremely rapid pace. Special measuring equipment is developed for them, and special mathematical algorithms are created to determine the general optical spectral characteristics of the atmosphere and the microphysical parameters of its aerosol component [1–3].

In work [4], the authors proposed a method (hereafter referred to as the Method) to recover the microphysical parameters of the multimode aerosol component in the atmosphere using spectral polarimetric measurements of the sky. It allows the number of main aerosol modes and some microphysical parameters of the engaged particles, as well as the spectral values of the general parameters of the gas-aerosol component in the atmosphere environment, to be determined. As a practical example of the Method application, a step-by-step process of recovery of atmospheric parameters using the data of measurements performed in [5] was considered. The characteristics of the main aerosol modes in the atmosphere recovered using the Method turned out close to the results obtained for the same observation position using modern measurements and the control algorithms of the AERONET network [6].

The aim of this work was to verify the efficiency of the Method algorithms at processing the spectral phase dependences of the sky linear polarization degree obtained in [7] and compare the recovered values of microphysical parameters for aerosol mode particles with the corresponding values obtained in [7] and with the values measured by the instruments and data processing algorithms of the AERONET network [8].

2. Main Part

In work [7], the results of radiation and polarimetric measurements of the sky above the position of the Cabauw Experimental Site for Atmospheric Research (CESAR Observatory), the Netherlands (51.971° N, 4.927° E) were reported. The measurements were carried out using the spectropolarimetric instrument groundSPEX, which was specially developed for this purpose. Using the measurement data obtained under clear sky conditions at 14:55 UTC on July 9, 2013, the spectral dependences of the brightness and the linear polarization degree (DoLP) of the sky on the light scattering angle in the main plane were plotted; see Fig. 1. The cited work also contains model curves calculated by means of algorithms developed for solving the inverse problem [9,10] using the parameter values of the aerosol component in the atmosphere that were recovered by the groundSPEX support group.

In this work, on the basis of the indicated spectral phase dependences of the sky DoLP, the microphysical parameters of particles in the coarse- (Coarse mode) and fine-dispersed (Fine mode) aerosol modes in the sky above the observation position of the CE-SAR Observatory at 14:55 UTC on July 9, 2013, were recovered. The following quantities were determined: the effective radius, r, and dispersion, σ^2 , of the normal logarithmic particle size distribution function and the real part of the complex refractive index of particles, n_r . For the DoLP parameter in the Coarse mode, the weighting coefficient coef₁ was fitted, which allowed us to correctly calculate the total DoLP value for the aerosol component in the atmosphere. In addition, the spectral values of the parameter describing the relative contribution of molecular scattering $\beta(\lambda)$ in the atmosphere at the light wavelengths $\lambda = 675$ and 870 nm were determined. The successive stages of this procedure were considered.

A qualitative analysis of the dependences shown in Fig. 1 showed that, despite the conditions of clear-sky measurements pointed out in [7], the absolute values of DoLP do not reach a value of 0.5 even in the short-wavelength interval of visible light. This obviously points to the presence of a substantial amount of aerosol in the air column during the measurements, which led to a considerable decrease in the observed

values of the sky DoLP and reduced the accuracy of the measurements, especially at small light scattering angles. In addition, the spectral differences in the observed dependences of the sky DoLP testify to a substantial effect of light scattering just at the particles of the fine-dispersed mode of atmospheric aerosol. The spectral dependences of the sky DoLP on the light scattering angle in the main plane were given in work [7] only as plots (see Fig. 1); therefore, they were digitized using the Graph2Digit 0.7.1b software [?]. For every scattering angle, the DoLP values were determined by averaging over the given vertical bars corresponding to the measurement error.

The parameters of the gas-aerosol environment of the atmosphere were recovered using the abovementioned Method. Here, we only briefly present its key features and formulas; one can read more about it in work [4]. The main idea of the Method is to represent the studied atmosphere as a homogeneous gas-aerosol model environment, whose DoLP spectral phase characteristics are determined by a specific set of microphysical parameters. The data of the phase measurements of the sky DoLP above the observation position were compared with the corresponding DoLP dependencies calculated for the model environment and the values of its microphysical parameters were fitted using the Method algorithm to achieve the best agreement for the indicated DoLP dependencies. For this purpose, the corresponding Root-Mean-Square Deviation (RMSD) functions were minimized; the plots of the latter clearly illustrate the process of finding the required parameter values. For the model gas-aerosol environment, the dependence $P(\alpha, \lambda)$ of the DoLP on the light scattering angle α and the wavelength λ has the form [12]

$$P(\alpha, \lambda) = \beta(\lambda)P_R(\alpha) + (1 - \beta(\lambda))P_a(\alpha, \lambda), \tag{1}$$

where $P_R(\alpha)$ is the gas component of DoLP, $P_a(\alpha, \lambda)$ is the aerosol component of DoLP, and

$$\beta(\lambda) = \frac{\sigma_R(\lambda)}{\sigma_R(\lambda) + \sigma_a(\lambda)} \equiv \frac{\tau_R(\lambda)}{\tau_R(\lambda) + \tau_a(\lambda)}$$
(2)

is the spectral value of the relative contribution of molecular scattering in the atmospheric environment. In formula (2), $\sigma_R(\lambda)$, $\sigma_a(\lambda)$, $\tau_R(\lambda)$, and $\tau_a(\lambda)$ are the volume scattering coefficients (σ) and the optical thicknesses (τ) of the gas (R) and aerosol (a) components in the atmosphere at the wavelength λ .

The gas contribution $P_R(\alpha)$ to the total sky DoLP depends on the light scattering angle and, according to the Method, takes into account a single scattering at the molecules of atmospheric gases (the Rayleigh scattering),

$$P_R(\alpha) = \sin^2(\alpha)/(1 + \cos^2(\alpha) + \delta/(1 - \delta)), \tag{3}$$

where δ is the depolarization index. For small gas optical thicknesses of the atmosphere ($\tau_R \ll 1$), the value of $P_R(\alpha)$ should reach maximum values of about 95% at scattering angles of 90° and 270°. Under real conditions, due to the depolarizing effect of multiple scattering, the observed value $P_R'(\alpha)$ is smaller, which is especially noticeable in the short-wavelength spectral interval of visible light.

According to the Method, to consifer the nature and the parameters of the particle size distribution function for the aerosol component of the atmospheric environment, the latter is represented as a combination of polydisperse systems of uniform spherical particles (the aerosol modes) characterized by a normal-logarithmic size distribution function. In particular, when using a model of the atmospheric environment containing two main aerosol modes, the total DoLP value of the aerosol component is determined by the expression

$$P_a(\alpha, \lambda) = \operatorname{coef}_1 \cdot P_{a1}(\alpha, \lambda, \rho_1, n_{r1}) + (1 - \operatorname{coef}_1) \times \times P_{a2}(\alpha, \lambda, \rho_2, n_{r2}),$$
(4)

where $P_{a1}(\alpha, \lambda, \rho_1, m_{r1})$ is the DoLP component formed by aerosol mode 1, $P_{a2}(\alpha, \lambda, \rho_2, m_{r2})$ is the same for mode 2, coef_1 is the weighting factor for mode 1, $\rho_{1,2} = 2\pi r_{1,2}/\lambda$ are the Mie parameters of modes 1 and 2, $r_{1,2}$ are the effective radia of aerosol particles of modes 1 and 2, $m_{1,2} = n_{r1,2} - j n_{i1,2}$ are the complex refractive indices (CRIs) of particles of modes 1 and 2, and $n_{r1,2}, n_{i1,2}$ are the spectral values of the real and imaginary, respectively, parts of the CRI of particles of modes 1 and 2. We emphasize that, in the algorithm of the Method, light absorption in the continuous spectrum is neglected because the vast majority of aerosol particles detected in the Earth's atmosphere are weakly absorbing [4].

Additionally, the Method uses the sky DoLP dependences measures under clear sky conditions, when the presence of even strongly light-absorbing aerosols in air is very low. To simulate the DoLP spectral

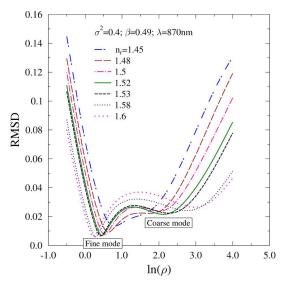


Fig. 2. RMSD dependencies on $\ln(\rho)$ for various n_r -values of model aerosol particles

phase dependences of light scattered in a gas-aerosol environment, we developed a set of special computer program codes that allow calculations with an accuracy of 10^{-8} .

2.1. Simulation procedure

The procedure for selecting microphysical parameters of aerosol particles was carried out at the wavelength $\lambda=870$ nm because the influence of multiple scattering on the sky polarization characteristics is insignificant in the long-wavelength interval of the visible light spectrum [12, 13].

2.1.1. Stage 1

The initial stage of data analysis was performed according to the method proposed in work [14]. Its results showed a general picture for the distribution of the main modes of the aerosol component in the atmosphere and narrowed the search interval for probable parameter values of their particles at the next stages. In Fig. 2, the plots of the RMSD function calculated in the framework of the model of the single-mode aerosol component in the atmosphere (coef₁ = 1) are shown. The calculations were performed in the widest possible interval of physically permissible values for the Mie parameter of aerosol particles (the expression $\ln(\rho)$ is used for it in this work) and for various values of the real part of their refractive index n_r . Recall that the minimum value of the RMSD

function corresponds to the best agreement between the calculated model values of DoLP and the experimentally determined values of the sky DoLP at all measured light scattering angles.

It should be noted that Fig. 2 also demonstrates the values of the parameters σ^2 and $\beta(870 \text{ nm})$, which were used in the calculations of the exhibited dependences. These as yet preliminary parameter values were selected from the intervals of their permissible values only after a few circles of estimation calculations, and their exact values were determined at the subsequent stages of the analysis.

Hence, the kit of the calculated RMSD dependences shown in Fig. 2, testifies to the presence of two main modes in the aerosol component in the atmosphere above the observation position. The domains of the probable existence of these modes are marked as "Coarse mode" and "Fine mode". The ratio between the minimum values of the RMSD function in these domains demonstrates that the Fine mode quantitatively exceeds the Coarse mode. Therefore, for further analysis, we determined the following intervals of probable values for the particle parameters: the parameter $n_r = 1.48 \div 1.53$ for both aerosol modes; the parameter $\ln(\rho) = 1.5 \div 3.0$ for the Coarse mode, and $\ln(\rho) = 0.1 \div 0.7$ for the Fine mode.

2.1.2. Stage 2

At the next stage, we fitted the parameters for the Coarse mode. Figure 3 demonstrates the dependences of the RMSD function calculated at the wavelength $\lambda = 870$ nm within the intervals of probable parameter values of particles in the Coarse mode and the parameter β_1 of the gas-aerosol environment (the notation β_1 reflects the application of the model of the single-mode aerosol component in the atmosphere).

As one can see from Fig. 3, a, the shape of the RMSD dependence on the parameter $\ln{(\rho_c)}$ and the position of its minimum are highly sensitive to relatively small value changes of the parameter β_1 . We successfully compensated this sensitivity by stepwise fitting the value of the parameter β_1 , which substantially narrowed the search interval of the Mi parameter values for the Coarse mode particles to $1.5 \div 2.5$ and allowed us, using simple averaging, to determine the most probable value $\ln{(\rho_c)} \approx 2.0$.

Figures 3, b–c illustrate the recovery procedure for other parameters of the Coarse mode particles and the refinement of the value of the parameter β_1 . We

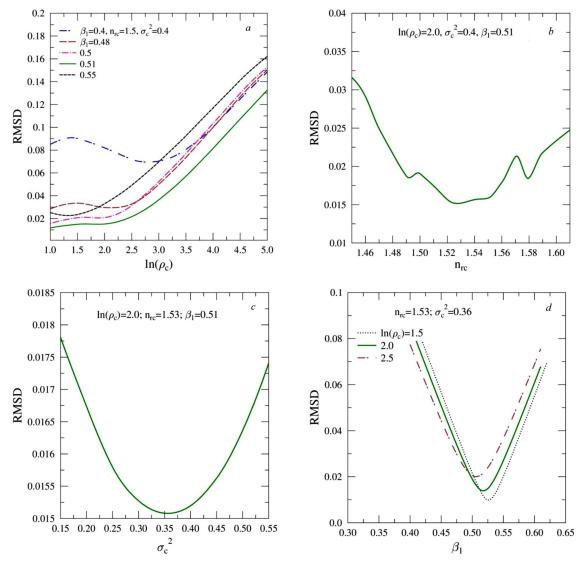


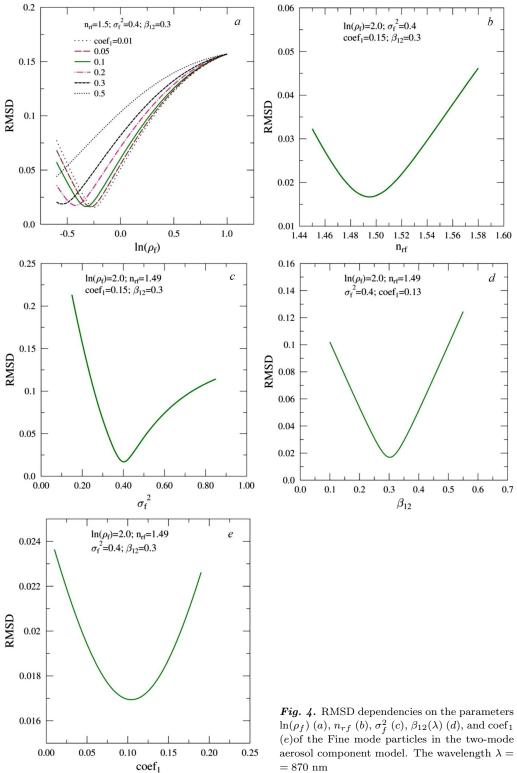
Fig. 3. RMSD dependencies on the parameters $\ln(\rho_c)$ (a), n_{rc} (b), σ^2 (c), and $\beta_1(\lambda)$ (d) of the Coarse mode particles of the single-mode aerosol component in the atmosphere. The wavelength $\lambda = 870$ nm

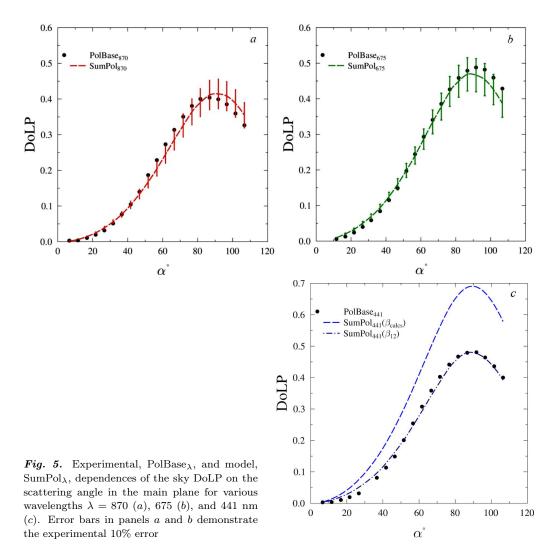
emphasize that when recovering the value of each subsequently sought parameter, the values of other parameters that have already been determined at the previous stages were used.

Figure 3, d demonstrates the value refinement process for the parameter β_1 . The RMSD dependences on this parameter plotted in this figure were calculated for the values of the parameter $\ln{(\rho_c)}$ within the interval indicated above. The solid curve shows the dependence calculated at the average value $\ln{(\rho_c)} = 2.0$ and has a minimum at $\beta_1 = 0.52$. Note that this

value of the parameter $\beta_1(870 \text{ nm})$ turned out considerably larger than the value $\beta_{\text{calc}}(870 \text{ nm}) = 0.295$ calculated according to Eq. (2) with the corresponding spectral values of the optical thickness for the gas and the aerosol components in the atmosphere taken from handbook [15].

Hence, besides the first indicator of the presence of the Fine mode in the atmosphere above the observation position, which was already detected in the qualitative analysis of Fig. 2, a significantly overestimated value of the parameter $\beta_1(870 \text{ nm})$ determined at this





analysis stage became another indicator of the presence of the Fine mode. Therefore, at the next stage, we recovered the parameters of the Fine mode particles and determined the quantitative ratio between the detected main modes in the aerosol component of the atmosphere.

2.1.3. Stage 3

In Fig. 4, the RMSD dependences on the Fine mode particle parameters and the parameter β_{12} (here the notation β_{12} reflects the use of the two-mode model for the aerosol component in the atmosphere) calculated for the wavelength $\lambda=870$ nm are depicted. According to the Method algorithm, the DoLP value of the aerosol component at this analy-

sis stage is formed by the parameter $DoLP_c$ of the Coarse mode and the parameter $DoLP_f$ of the Fine mode, and taking into account the weighting coefficient $coef_1$ of the Coarse mode (4).

As a result of simulation, from the position of the RMSD function minima, we determined the values of the parameters of the Fine mode particles and the parameter β_{12} . Note that besides the dependence of the RMSD function on the parameter $\ln{(\rho_f)}$, Fig. 4, a also demonstrates how the coefficient coef_1 modifies this dependence. The calculations show that the variation of coef_1 within an interval of $0.01 \div 0.2$ affects very weakly on the absolute value of the RMSD function minimum; at the same time, it substantially changes the fitted value of the parameter $\ln{(\rho_f)}$. The-

refore, using the already determined parameters of the Fine mode particles, we refined the value of this weighting coefficient, which gave us the value $coef_1 \approx 0.1$; see Fig. 4, e.

Thus, the quantitative ratio between the Coarse and Fine modes in the aerosol component of the atmosphere at the observation position is approximately equal to 1:9. This result expectedly confirmed our initial conclusions concerning a substantial quantitative excess of the Fine aerosol mode over the Coarse one.

3. Discussion

Figure 5 demonstrates the spectral dependences of the sky DoLP on the light scattering angle α in the main plane. The figure exhibits the experimental PolBase $_{\lambda}$ dependences averaged over the data presented in Fig. 1 and the model SumPol $_{\lambda}$ dependences calculated using the recovered parameters of the gasaerosol environment in the atmosphere.

According to the conclusions made in work [12], at wavelengths shorter than 600 nm, the calculation accuracy of the sky polarization characteristics in the single scattering model does not exceed 10%. The vertical bars in Figs. 5, a and b mark the intervals

Table 1. Recovered values of the mode parameters of aerosol component in the atmosphere

Para- meters	Aerosol modes					
	Coarse mode		Fine mode			
	Data source					
	This work	AERONET/ groundSPEX [7]	This work	AERONET/ groundSPEX [7]		
$r, \mu m$	0.9	$(1.5-4.8)/ \ (0.8-6.5)$	0.1-0.11	0.13-0.2/ 0.12-0.3		
σ^2	0.36	_	0.4	_		
n_r	1.53	(1.4-1.63)/ (1.42-1.58)	1.49	(1.4-1.63)/ (1.42-1.58)		
$coef_1$	0.1–0.11	_	_	_		

Table 2. Spectral values of the parameter $\beta(\lambda)$

Wavelength, λ , nm	870	675	441
$\beta_{12}(\lambda)$, this work	0.3	0.46	0.53
$\beta_{\rm calc}\lambda$, handbook [15]	0.295	0.46	0.74

of the 10% error for the calculated SumPol $_{\lambda}$ dependences. As one can see, at wavelengths of 870 and 675 nm, the shapes of the model curves are close to the experimental ones and intersect with the averaged measurement data within the indicated calculation accuracy. Note also that according to the data in [7, Table 1], the measurement accuracy of the PolBase $_{\lambda}$ dependences is determined by the pairs of systematic and random errors, and for most of the examined light scattering angles, it considerably exceeds the 10% value. As a result, the intersection of the intervals of the measured and simulated phase dependences of the sky DoLP becomes larger so that the recovered values of the parameters obtained in this work can be considered close to the real ones.

Figure 5, c illustrates the results obtained for the wavelength $\lambda=441$ nm. Here, the SumPol $_{\lambda}$ dependences are shown that were calculated at the parameter value $\beta_{12}(441 \text{ nm}) = 0.53$ fitted using the Method algorithm and the "reference" value $\beta_{\text{calc}}(441 \text{ nm}) = 0.74$. According to the conclusions made in work [12], owing to the influence of multiple light scattering at short wavelengths, the calculation error of the sky DoLP value in the single-scattering Rayleigh model increases substantially; therefore, the 10% error interval cannot be used at longer wavelengths. As one can see, the dependence SumPol $_{\lambda}(\beta_{12})$ is much closer to the experimental PolBase $_{\lambda}$ curve than the dependence SumPol $_{\lambda}(\beta_{\text{calc}})$. The origin of this discrepancy will be considered below.

Table 1 contains the values of the recovered parameters for two main modes of the aerosol component in the sky above the position of the CESAR Observatory at 14:55 UTC on July 9, 2013, as well as the value intervals for the corresponding parameters of aerosol modes. The latter were determined from multi-day observations at this position and their processing in the AERONET network, and from the measurement results of the groundSPEX instrument [7]. As one can see, the parameters of aerosol modes recovered in this work do not go beyond the value intervals determined by the AERONET and groundSPEX algorithms. Note, however, that the effective particle radius values fitted for both aerosol modes are consistent with only the minimum values in the indicated intervals.

Table 2 quotes the spectral values of the parameter describing the relative contribution of molecular scattering in the atmosphere. They were determined

from the spectral phase dependences $\beta_{12}(\lambda)$ of the sky DoLP and calculated using the reference spectral values for the optical thickness of the gas and aerosol components in Earth's atmosphere $\beta_{\rm calc}(\lambda)$ [15]. The spectral values of the parameters fitted using the Method exactly correspond to the reference values $\beta_{12}(870 \text{ nm}) = \beta_{\rm calc}(870 \text{ nm}) = 0.3$ and $\beta_{12}(675 \text{ nm}) = \beta_{\rm calc}(675 \text{ nm}) = 0.46$, which testifies to the reliability of the recovered parameter values for the aerosol component.

At the same time, the fitted value of the parameter $\beta_{12}(441 \text{ nm}) = 0.53$ is substantially smaller than the "reference" value $\beta_{\text{calc}}(441 \text{ nm}) = 0.74$. This result means that the value of the parameter DoLP of the gas component calculated in the framework of the single light scattering model is significantly overestimated in comparison with its real value.

As another origin of this discrepancy, we consider the availability of a substantial amount of aerosol in the air above the observation position at the time of measurement; this circumstance was already pointed out above, when making the general qualitative analysis of the input data. It is obvious that under such conditions, the influence of multiple light scattering on the sky polarization characteristics increases substantially, and this fact is especially pronounced in the short-wavelength spectral interval. This assumption is confirmed by conclusions made in work [12] with respect to the measurement and calculation results obtained for the sky polarization parameters in the framework of the single Rayleigh scattering model at wavelengths shorter than 600 nm.

4. Conclusion

The efficiency of the Method proposed for the recovery of the microphysical parameters of the multimode aerosol component in the atmosphere has been demonstrated by analyzing the data from the spectral polarimetric measurements of the sky over the CE-SAR Observatory, the Netherlands, using the ground-SPEX device. The correctness of the recovered values of the microphysical parameters of the main aerosol modes is confirmed by the closeness of the results of the spectral phase measurements of the sky linear polarization degree over the observation position and the dependences of this parameter calculated for the model gas-aerosol environment. The aerosol particle parameters reconstructed in this work are consis-

tent with the intervals of their probable values determined in the AERONET network and calculated by the spectropolarimeter groundSPEX support group.

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О.С. Овсак, В.М. Ващенко, А.П. Відьмаченко ВІДНОВЛЕННЯ ПАРАМЕТРІВ ГАЗОВО-АЕРОЗОЛЬНОГО СЕРЕДОВИЩА АТМОСФЕРИ НАД ПОЗИЦІЄЮ CESAR OBSERVATORY, НІДЕРЛАНДИ

З використанням раніше запропонованого авторами Методу відновлення параметрів багатомодової аерозольної скла-

дової атмосфери, проаналізовано дані поляризаційних вимірювань неба над позицією CESAR Observatory, Нідерланди. В атмосфері над позицією виявлена присутність двох аерозольних мод з нормально-логарифмічним розподілом частинок за розмірами й відновлено низку їх мікрофізичних параметрів. Для грубодисперсної моди визначено дійсну частину показника заломлення $n_r = 1,53$, ефективний радіус частинок r = 0.9 мкм, їх дисперсія $\sigma^2 = 0.36$, ваговий коефіцієнт цієї моди у загальній величині ступеню лінійної поляризації аерозольної суміші $coef_1 = 0,1$. Для дрібнодисперсної моди $n_r = 1,49, r = 0,11$ мкм, $\sigma^2 = 0,4$. Кількісне співвідношення вказаних аерозольних мод у повітрі над позицією спостереження складало приблизно 1:9. Визначено спектральні величини відносного вкладу газового розсіяння: $\beta(870\,$ нм $)=0,3\,$ і $\beta(675\,$ нм)=0,46. Відмічено некоректність використання моделі однократного розсіювання Релея у короткохвильовій ділянці видимого спектру світла та при значній насиченості повітря аерозолями. Продемонстровано суттєвий вплив багатократного розсіювання світла на результати поляриметричних вимірювань на довжині хвилі 441 нм за вказаних атмосферних умов проведення спостережень неба.

K лючові с лова: земна атмосфера, дистанційні вимірювання, ступінь лінійної поляризації, аерозоль, відновлення мікрофізичних параметрів.