ISSN 0001-4338, Izvestiya, Atmospheric and Oceanic Physics, 2006, Vol. 42, No. 6, pp. 752–764. © Pleiades Publishing, Inc., 2006.
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Analysis of Solutions to the Inverse Problem on the Retrieval of the Microstructure of Stratospheric Aerosol from Satellite Measurements

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Received July 7, 2005; in final form, October 25, 2005

Abstract—A statistical ensemble of microphysical parameters of the background stratospheric aerosol at altitudes of 15 to 30 km is modeled on the basis of experimental data. The aerosol attenuation coefficients (AACs) in the wavelength range 0.38–16.3 μm are calculated for all realizations of the ensemble by algorithms of the Mie theory. Analysis of correlations between the AACs and the microphysical parameters indicate that the AAC correlates most strongly with the total volume V and area S of all particles. The errors of determining the microphysical parameters from AAC measurements are analyzed via the method of linear regression. It is shown that, if the AAC is measured with an error of 5%, the errors of determining both the particle size distribution (PSD) for particles with sizes of 0.4 to 4 μm and the parameter S are an order of magnitude smaller than the prior uncertainty, whereas the error of determining V is two orders of magnitude smaller than the prior uncertainty. Schemes of AAC measurements with the SAGE III, ISAMS, CLAES, HALOE instruments and an IR interferometer in the visible and IR regions are discussed. It is shown that combining the schemes makes it possible to extend the range of particle sizes for which the PSD is retrieved with a satisfactory accuracy and to increase the accuracy of determining S and V substantially and the accuracy of determining the total number of particles N_{opt} to a lesser extent. Examples of interpreting AAC measurements carried out simultaneously with the SAGE III and HALOE instruments within the same spatial region are presented. A systematic discrepancy between vertical profiles of S and V obtained from SAGE III and HALOE measurements is revealed.

DOI: 10.1134/S0001433806060090

1. INTRODUCTION

Microphysical parameters of stratospheric aerosol determine radiative characteristics of the atmosphere and cloud-formation processes to a great extent and affect the ozone content in the stratosphere [1]. For this reason, numerous studies of stratospheric-aerosol characteristics have been performed on the basis of local (see, for example, [2]) and satellite measurements on regional and global scales.

Satellite measurements of atmospheric transparency on slant paths (during risings or settings of the Sun, Moon, or stars) make it possible to determine the spectral–altitude aerosol attenuation coefficients (AACs) $\alpha(\lambda, z)$. Such data were obtained in the visible and near IR regions of the spectrum in the experiments with the SAM II, SAGE I, II, III, POAM II, III, Ozon–Mir, and GOMOS instruments [3–8]. Similar measurements were performed in the IR region of the spectrum (ATMOS, HALOE instruments [9–11]). Satellite instrumental measurements of the emission of the planet's limb in the IR region of the spectrum (CLAES, ISAMS) likewise allow one to obtain the aerosol attenuation (absorption) coefficients in different spectral instrument channels [12, 13].

All these data are used in the following to determine different microphysical parameters of both stratospheric aerosol and polar stratospheric clouds (PSCs) on the basis of different methods elaborated for solving inverse problems. Note that such optical methods of studying the microstructure of atmospheric aerosol are widely used in ground-based measurements (AERONET network [14]). The pioneering studies in this field were carried out by K.S. Shifrin and A.Ya. Perel'man in the 1950s (see, for example, [15, 16]). It should be emphasized that ground-based methods yield integral (over the entire atmospheric thickness) aerosol characteristics, whereas satellite measurements of atmospheric transparency on slant paths (or of the thermal radiation of the planet's limb) yield information about vertical profiles of aerosol characteristics.

As a rule, the inverse problem under consideration is formulated on the basis of the known expression for

the AAC for polydisperse aerosol (model of spherical homogeneous particles):

$$a(\lambda, z) = \int_0^\infty \pi r^2 Q_e(r, \lambda, m(\lambda, z)) n(r, z) dr, \qquad (1)$$

where $Q_e(r, \lambda, m(\lambda, z))$ is the Mie factor of attenuation efficiency of an aerosol particle with radius r and complex refractive index (CRI) of its matter $m(\lambda, z)$, $n(r, z)$ is the particle size distribution (PSD), λ is the wavelength, and z is the altitude in the atmosphere.

In a general case, Eq. (1) involves two unknowns: the PSD and the CRI. However, in most cases, it is assumed for the visible region of the spectrum that the CRI is known, for example, from previous experimental studies. Sometimes, the CRI for the background stratospheric aerosol is calculated with the corresponding models of its formation and from independent measurements of temperature, pressure, and water-vapor content [17].

The informativeness of AAC measurements in different spectral regions with respect to the PSD was measured by various methods; however, as a rule, semiquantitative methods (for example, comparison of the efficiency factors $Q_e(r, \lambda, m(\lambda, z))$ were used for different wavelengths [13, 18, 19]. Those studies have demonstrated their rather low informativeness and, as a consequence, the necessity of invoking a substantial amount of additional prior information to solve the inverse problem. As in the solution of other inverse problems of atmospheric optics, prior information about the PSD may be of different types—boundedness or smoothness of the desired function, its positiveness, belonging to an empirical statistical ensemble, etc. A specific analytic form of the PSD (for example, a unimodal or bimodal log-normal distribution) is often used as prior information [10].

In studies devoted to the use of IR measurements of the AAC, data of numerous local measurements of the PSD and its parameters are frequently employed as prior information [2]. For example, 774 PSD measurements performed with aerostat optical sensors from June 1991 to April 1994 were used in [19]. On the basis of those measurements in the period of a volcanic and a postvolcanic stratosphere, simple empirical relationships were established between the integral area of particles S and their integral volume V as well as the average radius of particles R_e. Using an approximate relation of the AAC in the IR region of the spectrum to the parameters R_e and V, the authors of [19] developed a simple method of determining the above parameters of the microstructure of stratospheric aerosol and PSCs from IR measurements of the absorption coefficients made with the HALOE, ISAMS, and CLAES instruments. The subsequent development of that method of solving the inverse problem and its use for analysis of satellite measurements were implemented in [9, 10, 12, 13].

Quantitative estimations of the accuracy and informativeness of AAC satellite IR measurements performed under different conditions in a wide spectral range are of interest if the possibility of determining the PSD and its integral parameters is concerned. Such studies are important because satellite and aerostat measurements of the AAC in the IR region of the spectrum are routinely conducted, including those with contemporary IR interferometers (see, for example, [11, 20]). The aforementioned studies [9, 10, 12, 13] related basically to a volcanic and a postvolcanic stratosphere. In this study, the informativeness of such measurements and the accuracy of the remote-sensing method under consideration are analyzed for the background-stratosphere conditions.

2. STATISTICAL MODEL OF THE BACKGROUND STRATOSPHERIC AEROSOL

When formulating a microphysical model of stratospheric aerosol, we used both experience in constructing earlier models [8, 21–24] and statistical characteristics of a PSD ensemble obtained from PSD long-term local measurements taken with optical particle counters mounted on aerostats [2]. In particular, an aerosol model was simplified relative to the model described in [21], which considered four fractions of stratospheric aerosol. The model described below suggests the presence of only one major fraction of the background aerosol, namely, a sulfuric acid liquid aerosol.

In addition, the experimental data obtained in [25–30] were used to extend the statistics of stratospheric PSDs during modeling of the variability of microphysical parameters. The statistics was extended for two reason:

(1) Our study was aimed at constructing a global model of stratospheric aerosol, whereas measurements [2] can be regarded as regional ones.

(2) The use of narrow (local) information for solving the inverse problem of determining the PSD causes the solution to depend only slightly on the results of AAC measurements (see, for example, [31]).

Out of all experimental data [2], we selected those related to the first (fine) fraction of stratospheric aerosol and measured in the years when the stratosphere was not influenced by volcanic emissions (1988–1991, 1996–2002). All in all, we considered 668 PSD measurements taken at different altitudes.

For modeling aerosol PSDs, we used a unimodal log-normal distribution

$$n(r) = \frac{N}{r\sqrt{2\pi}\ln\sigma} \exp\left[-\frac{\ln^2(r/r_g)}{2\ln^2\sigma}\right], \qquad (2)$$

where N is the total number of particles, r_g is the median radius, and σ is the width of the distribution.

When constructing PDS realizations $n(r, z)$ for our ensemble, we specified the average profiles of the parameters of distribution (2) (N and r_g) and their rms deviations (RMSDs). Deviations of these parameters from their average profiles at different altitudes must satisfy a log-normal multidimensional distribution, which is specified by the average profile of the logarithm and the model correlation matrix $\mathbf{D} = \{d_{i,j}\}$ whose elements are defined as follows:

$$d_{i,j} = \Sigma_i \Sigma_j \exp\{-|z_i - z_j|/r_c\}. \quad (3)$$

Here, i and j are the indices that designate altitude levels, Σ is the rms variation, z is the altitude, and r_c is the correlation radius. The correlation radius in altitudes is taken equal to 5 km. The average profiles of the total number of particles N and the median radius r_g were taken from [2]. The RMSD at all altitudes was set equal to 0.7 for N (in an experimental ensemble, it assumed 0.47–0.82 depending on the altitude) and 0.55 for r_g (in an experimental ensemble, it assumed 0.32–0.67). As a result of analysis of measurements [2], we determined a statistical relation between the median radius and the width of the distribution of log-normal PSDs. During modeling, this relation was specified as a piecewise linear function such that the deviation of the distribution's width σ from this function was 0.25.

On the basis of analysis of the variability of the PSD parameters [2, 25–30], we imposed the following restrictions on the parameters: the range of variation of the median radius was 0.01–0.49 and the range of variation of the distribution's width was 1.2–3.5.

In order to determine the chemical composition of the background stratospheric aerosol, i.e., the percentage content of sulfuric acid H_2SO_4 and, consequently, the CRI of particles (which depends in the IR spectral region on the stratospheric temperature and humidity), and to calculate aerosol optical parameters, we included information about thermodynamic parameters (pressure, temperature, water-vapor content) into the ensemble of stratospheric states. Variations in the profiles of these parameters were modeled with the well-known AGFL-86 models [32]. Thus, both the PSD and the CRI were varied in the formulated statistical microphysical model of stratospheric aerosols. This peculiarity distinguishes our approach from the studies in which the CRI of particles was assumed to be known (see, for example, [36]).

A global ensemble must include maximum variations in parameters. Therefore, the statistical ensemble of realizations of the atmospheric temperature and humidity was based on five models: for tropical latitudes, middle latitudes (summer, winter), and subarctic latitudes (summer, winter) [32]. Natural variations were set equal to 10 K for temperature for all climatic models and 30% of the average relative humidity at the altitude under consideration for humidity. The correlation radius r_c (see formula (3)) was taken as 10 km for temperature and 5 km for humidity. The generated ensemble of parameters of the atmospheric state consisted of 1200 realizations of profiles of pressure, temperature, humidity, and PSD and CRI parameters at each of 40 altitudes from 10 to 30 km.

Realizations at four altitudes (15, 20, 25, 30 km) were considered for numerical analysis of the informativeness of AAC measurements and the accuracy of solving the inverse problem on the retrieval of the PSD and CRI (percentage of sulfuric acid Wt) from optical measurements in the IR spectral region. Thus, the ensemble of initial data consisted of 4800 realizations of parameters of stratospheric aerosol. Comparison of variations in different parameters of the PSD and Wt in the modeled ensemble with similar experimental values [2] has shown their good quantitative agreement. Moreover, our ensemble turned out to be wider in most characteristics than the data of local measurements [2] and other experimental information.

3. STATISTICAL CHARACTERISTICS OF MICROPHYSICAL AND OPTICAL PARAMETERS OF STRATOSPHERIC AEROSOL

On the basis of the generated ensemble of PSD and CRI realizations and AAC calculations, we calculated the averages and covariance matrices of aerosol microphysical and optical characteristics for a global model of the stratospheric state at altitudes of 15 to 30 km. The following parameters were included by us into the aggregate covariance matrix: the PSD, its moments (total number N_{opt}, surface area S, and volume V of optically active particles), the effective radius R_{eff}, PSD parameters (total number of particles, median radius N, and distribution width σ), and the percentage of sulfuric acid in the atmosphere Wt. The PSD was specified on 17 ranges of particle sizes (bins) from 0.01 to 10 μm. The above functionals $n(r)$ and the effective radius of particles R_{eff} were calculated from the formulas

$$N_{opt} = \int_{0.01}^{10} n(r)dr, \quad S = 4\int_{0.01}^{10} \pi r^2 n(r)dr,$$
$$V = \frac{4}{3}\int_{0.01}^{10} \pi r^3 n(r)dr, \quad R_{eff} = \frac{3/4\pi V}{1/4\pi S}. \quad (4)$$

The limits of integration with respect to particle sizes correspond to the range of particles optically active in the spectral range under consideration. If

$D_{\text{AAC AAC}}$	$D_{\text{AAC PSD}}$	$D_{\text{AAC XX}}$
$D_{\text{PSD AAC}}$	$D_{\text{PSD PSD}}$	$D_{\text{XX AAC}}$
$D_{\text{XX AAC}}$	$D_{\text{XX PSD}}$	$D_{\text{XX XX}}$

Fig. 1. Schematic representation of the composite covariance matrix of aerosol parameters (block D_{XX} includes all parameters except for PSD and AAC).

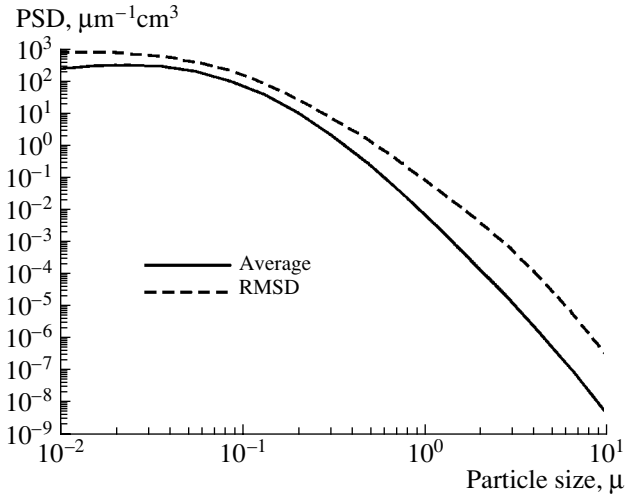

Fig. 2. PSD average and RMSD in an ensemble of realizations of stratospheric aerosol (15–30 km).

wider limits of integration are taken so that all PSDs were contained within these limits, differences in the ensemble under consideration are observed only for the total number of aerosol particles; therefore, the index opt is omitted for other moments. The aggregate covariance matrix also includes AACs (in 2201 spectral channels from 1.92 to 12.5 μm, in accordance with the measuring scheme of the MK-IV interferometer [18, 20]).

The elements of the covariance matrix of aerosol parameters **D** contain information about their statistical characteristics and were calculated from the formula

$$d_{ij} = \frac{1}{n-1}\sum_{n}(x_i - x_a)(x_j - x_a). \quad (5)$$

Here, n is the number of realizations (4800 in our case), x are the elements of the composite vector of aerosol parameters **x**, and the index a relates to the ensemble average. Since the composite vector **x** consists of many parameters, the composite covariance matrix **D** can be represented as a partitioned matrix each block of which relates to a definite parameter.

Figure 1 schematizes the composite matrix **D**. Each block along the diagonal contains the covariance matrix of an individual parameter (AAC, PSD, vector of the other parameters). Nondiagonal blocks of the composite matrix represent cross covariances between these parameters and are used in the regression approach to the solution of the inverse problem ($\mathbf{D}_{xy}$ blocks, see below). Analysis of individual diagonal blocks makes it possible to examine statistical characteristics of different parameters of stratospheric aerosol.

Figure 2 presents averages and RMSDs for the PSD in the range of particle sizes 0.01–10 μm for altitudes of 15 to 30 km. The peak of the average PSD (about 360 cm^{-3} μm^{-1}) falls within the range of particle sizes from 0.02 to 0.03 μm; i.e., fine particles predominate in the ensemble under consideration. (Recall that we consider the background atmospheric aerosol.) The peak of the absolute variation in the model of the number of particles in bins falls within the range of particles 0.01–0.02 μm and reaches 900 to 930 cm^{-3} μm^{-1} (more than 300% of relative variation). Starting with particle sizes of 0.07 μm toward larger sizes, the relative variation of the number of particles in bins increases, reaching more than 6000% for particles with a size of 10 μm. Thus, it can be seen that the variation of the PSD is sufficiently large in the statistical model under consideration.

Analysis of the variability of PSD integral parameters indicates that the variations of N_{opt}, S, and V are 175% of the average (44.5 from 25.5 cm^{-3}), 219% of the average (5.57 from 2.54 $\mu\text{m}^2/\text{cm}^3$), and 463% of the average (0.88 from 0.19 $\mu\text{m}^3/\text{cm}^3$), respectively. The increase in the relative variation with increasing order of the PSD moment is due to the increasing influence of larger-size particles, which show larger relative variations. The variations of the PSD parameters are 71% (from 0.05 to 0.07 μm) for the radius r_g and 16% (from 0.29 to 1.79) for the distribution's width σ. The variation of Wt is 7.4% at an average of 74.8% (i.e., about 10% of the average). Note that the variation of R_{eff} in the modeled ensemble is about 50% (0.08 from 0.15 μm). The averages of R_{eff} are small, a result that is characteristic of the background stratospheric aerosol. Thus, the predominant sizes of particles are substantially smaller than the wavelengths in the spectral range of AAC measurements

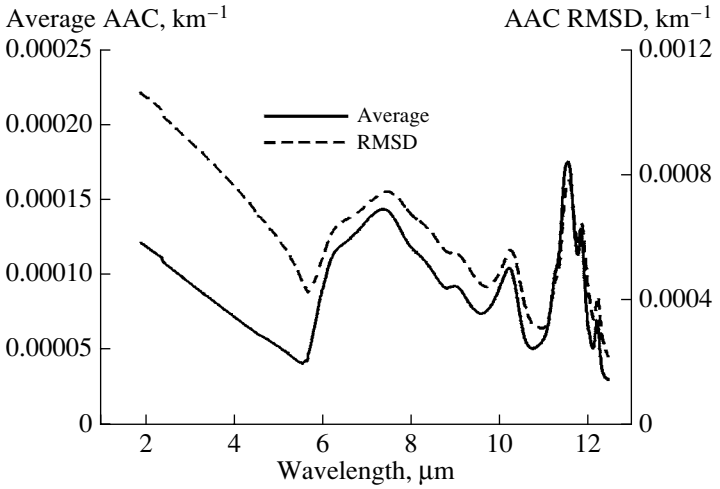

Fig. 3. AAC average and RMSD in an ensemble of realizations of stratospheric aerosol (15–30 km).

(1.9–12.5 μm). Thus, the averages and RMSDs obtained for different parameters in our model clearly characterize the wide range of the modeled ensemble of microphysical characteristics of the background stratospheric aerosol.

Since we are considering a number of microphysical parameters, including the composition of aerosol, it is of interest to examine the correlations between these parameters calculated from cross-covariance matrices (5). Analysis of the correlation coefficient (CC) between PSD parameters (r_g and σ), PSD functionals (N_{opt}, S, and V), and the percentage of H_2SO_4 in the atmosphere Wt has shown that significant correlations (CC > 0.5) are observed between the following characteristics: S and N_{opt} (CC = 0.53), S and V (CC = 0.8), σ and r_g (CC = –0.53), and R_{eff} and r_g (CC = 0.71). It should be noted that, in the ensemble under consideration, there are virtually no correlations between V and Wt, V and σ, R_{eff} and Wt, and R_{eff} and N_{opt}.

Figure 3 shows the spectral behaviors of both the ensemble-average AAC (left-hand ordinate axis) and its absolute RMSD (right-hand ordinate axis) in the wavelength range 1.92–12.5 μm that are calculated for the altitude range 15–30 km. As can be seen from the figure, in the wave-number range under consideration, the spectral behaviors of both the AAC and its RMSD change considerably and local maxima of the AAC, which are due to the spectral behavior of the imaginary part of the CRI, are observed in individual spectral ranges. The maximum values of the AAC fall within channels near 7.5 and 11.5 μm, and the peak of AAC absolute variation lies in the short-wavelength region of the spectrum.

4. INFORMATIVENESS OF AAC MEASUREMENTS IN THE IR REGION OF THE SPECTRUM AND ERRORS IN RETRIEVAL OF MICROPHYSICAL CHARACTERISTICS OF STRATOSPHERIC AEROSOL

From AAC values (see formula (1)) obtained by us for the generated ensemble of PSDs, we calculated the eigenvectors of the AAC covariance matrix and the related eigenvalues. On the basis of analysis of the eigenvectors and the expansion of the AAC spectral behavior in terms of these vectors for all ensemble realizations, we estimated the number of independent components of information in AAC measurements at different measurement errors (according to V.P. Kozlov [33]). Note that only if the accuracy of AAC measurements is high (10%) over the entire IR region under consideration (1.92–12.5 μm) can one obtain five independent components of information described by five eigenvectors of the AAC covariance matrix; the exact retrieval of the parameters of log-normal distribution (2) is not concerned here, because, as follows from experimental data [2], strong correlations may exist between these parameters.

In order to qualitatively analyze the possibility of solving the inverse problem of retrieving aerosol microphysics from AAC spectral measurements in the IR region of the spectrum, it is useful to examine statistical relations between the parameters. We studied correlations of the AAC with PSD parameters (r_g, σ), PSD moments (N_{opt}, S, V), and R_{eff} and Wt. Figure 4 shows the CCs between the AAC and the above parameters. It is seen that, for most aerosol parameters, their CCs with the AAC in the IR region of the spectrum are low in the ensembles under consideration. For example, the CCs of the percentage of sulfuric acid, N_{opt}, and σ with the AAC are no greater than

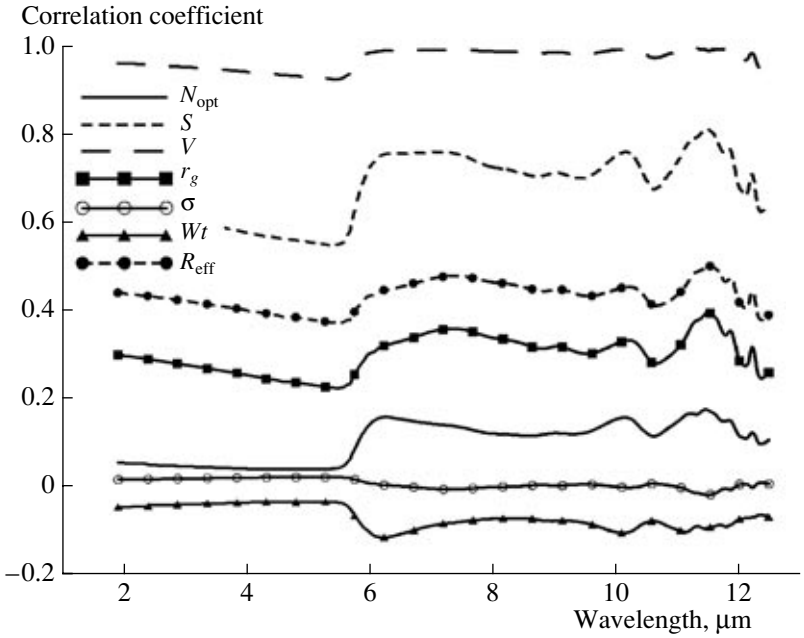

Fig. 4. Spectral behavior of the coefficient of correlation between the AAC and different aerosol parameters in an ensemble of realizations of stratospheric aerosol.

0.2 in absolute value. The CC between the median radius r_g and the AAC lies within 0.2–0.4, and the CC between R_{eff} and the AAC lies within 0.4–0.5. For S and V, their CCs with the AAC are 0.6 to 0.8 and 0.9 to 1.0, respectively. The greater these CCs, the greater the AACs themselves. Figure 4 also shows that the CCs increase during switching from 5 to 6 μm. The CCs between the AAC and the PSD moments and between the AAC and the aerosol composition have local maxima at wavelengths near 10 and 11.5 μm. The relatively high correlations found between the AAC and both S and V are in good agreement with the results of studies [11, 13, 19] obtained for a turbid atmosphere and are an additional justification that the simple method proposed in those studies to solve the inverse problem for the background stratosphere is applicable in the IR region of the spectrum.

Using the AAC spectral covariance matrix and the cross-covariance matrix between the AAC and aerosol microphysical parameters, we evaluated the errors in retrieval of the aerosol microphysical parameters (inverse-problem solution) by the well-known regression formula

$$\mathbf{x} = \mathbf{x}_a + \mathbf{D}_{xy}(\mathbf{D}_{yy} + \mathbf{I}\varepsilon^2)^{-1}(\mathbf{y} - \mathbf{y}_a). \quad (6)$$

Here, $\mathbf{x}$ is the vector of the desired microphysical parameters (PSD, its moments, etc.), $\mathbf{y}$ is the vector of AAC measurements, $\mathbf{D}$ is the corresponding blocks of the composite cross-covariance matrix (Fig. 1), $\mathbf{I}$ is the unit matrix, and ε is the error in measuring the AAC. The index a relates to an ensemble average. This formula was used in numerical experiments based on a closed scheme to retrieve the PSD and its parameters for 4800 realizations of the state of the background stratospheric aerosol. For each realization, the PSD and its parameters were calculated from regression formula (6) with account for random measurement errors (5–25%) added to the calculated AAC values. The calculated PSD and its parameters were compared to the initial "exact" values. After all realizations were examined, the ensemble rms characteristics of retrieval errors were calculated.

PSD Determination

Figure 5 presents the relative (upper panel) and absolute (lower panel) errors in PSD retrieval from the AACs known in 2201 spectral channels of the IR spectral region with errors of 5 and 25% (see [18, 20] for the scheme of MK-IV interferometer measurements). This figure also shows the prior variability of the PSD ensemble under consideration. It can be seen that the minimum errors in determining the PSD from AAC measurements (smaller than 100%) are obtained for particles with sizes from 0.1 to 3–5 μm. The prior uncertainty in the PSD for particles with sizes of 0.4 to 4 μm at a 5% error in AAC measurements is decreased from 500–5000% to 20–50%. For particles with sizes of 2 to 4 μm, the absolute error in determining the PSD is decreased by two orders of magnitude. For particles with sizes greater than 4 μm, the relative retrieval errors increase. Note that the informativeness obtained via comparison of the prior uncertainty and the error in the solution of the inverse problem for the

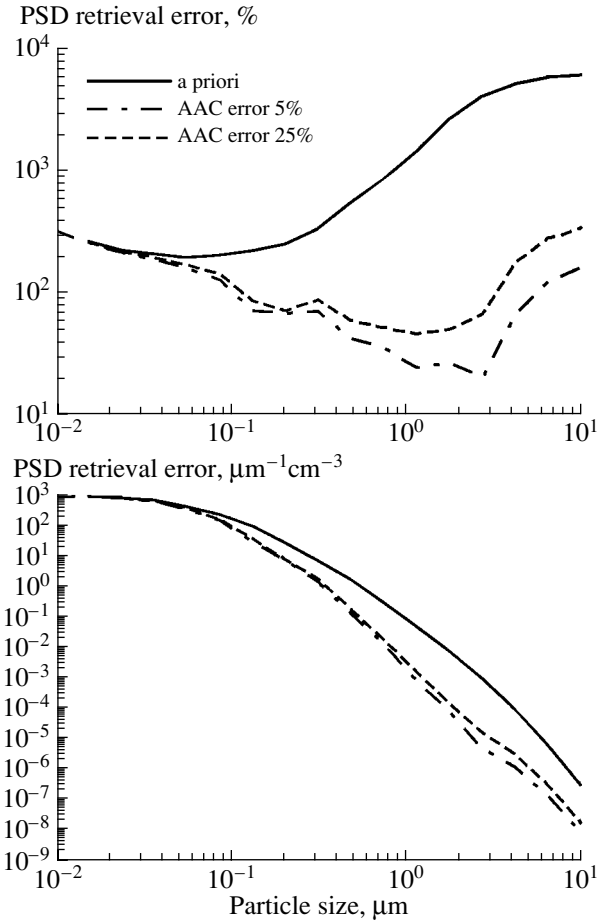

Fig. 5. Relative (above) and absolute (below) errors in PSD retrieval from AAC values specified with errors of 5 and 25% in the wavelength range 1.92–12.5 μm.

PSD from AAC measurements is as yet high for particles with such sizes. On the other hand, the number of particles with sizes close to 10 μm in most realizations of the ensemble modeled for the background stratospheric aerosol is so small (see, for example, Fig. 1) that AAC measurements in the IR region of the spectrum virtually do not introduce any additional information about the PSD.

From analysis of the total (over all particle sizes) errors in PSD retrieval, it follows that, in many cases, large total errors are due to large errors in PSD retrieval in small bins. This inference can be important when the retrieved PSD is used to determine the total number of particles. For large particles, which dominate in determining such integral parameters of the PSD as *S* and *V*, PSD is usually retrieved with a higher accuracy.

Determination of PSD Parameters

Consider the problem of determining the parameters r_g and σ of the PSD, its moments (*N*, *S*, *V*), and R_{eff} and *Wt*. Analysis of correlations (see Fig. 4) of these quantities with the AAC on the modeled ensemble of stratospheric aerosol has shown that significant correlations occur only between the AAC and *S* and between the AAC and *V*.

Table 1 presents data on the prior variability of the above parameters in the ensemble under consideration and on the errors in determining these parameters by the regression method from the AAC values measured in 2201 channels (1.92–12.5 μm) with errors of 5 and 25%. Analysis of Table 1 shows that, at a 5% error in specifying the AAC, the errors in determining *S* and *V* are respectively 7.6 and 293 times smaller than the prior uncertainties (0.73 versus 5.57 $\mu m^2/cm^3$ and 0.003 versus 0.88 $\mu m^2/cm^3$). The errors in determining the other parameters (r_g, σ, *N*, R_{eff}, *Wt*) are decreased only by 5 to 30% as compared to the prior uncertainties.

Analysis of Different Schemes of AAC Satellite Measurements

Satellite measurements of the AAC were performed with a number of instruments in both the visible and IR regions of the spectrum. Onboard the UARS satellite, for example, there were several instruments that measured optical characteristics of stratospheric aerosol in the IR region of the spectrum: CLAES, ISAMS, and HALOE [13, 34]. Measurements with the HALOE instruments are being continued at the present time. It is of interest to examine the potentials of different spectral schemes of measurements and their informativeness in combined experiments on determining different microphysical parameters of stratospheric aerosol. For this purpose, we

Table 1. Absolute error in determining microphysical parameters from AAC values measured with errors of 5 and 25% in the range 1.92–12.5 μm

	N_{opt}, cm^{-3}	*S*, $\mu m^2/cm^3$	*V*, $\mu m^3/cm^3$	r_g, μm	σ	*Wt*, %	R_{eff}, μm
a priori	44.4	5.57	0.88	0.046	0.29	7.39	0.082
5%	33.7	0.73	0.003	0.038	0.28	6.56	0.065
25%	35.8	0.90	0.014	0.038	0.28	6.67	0.066

will consider an example of modeling spectral data of AAC measurements with the SAGE III instrument (visible and near IR regions); the MK-IV and ATMOS interferometers (IR region); and the HALOE, CLAES, and ISAMS instruments.

Table 2 lists the spectral channels used for AAC measurements with the instruments under consideration. Recall that the region of measurements of the IR interferometers includes 2201 channels in the range from 1.92 to 12.5 μm. In addition to the potentials of individual instruments, we will examine two combined schemes: (1) IR + SAGE III and (2) measurements in all aerosol channels of the SAGE III, HALOE, CLAESS, and ISAMS instruments.

Figure 6 presents the relative error in PSD retrieval (in 17 bins from 0.01 to 10 μm) from the AACs measured with an error of 25%. The results are presented for AAC measurements with each of the four instruments (SAGE III, ISAMS, CLAES, HALOE) and for the experiment taking into account simultaneous measurements in all channels. The figure also presents the prior variability of the PSD ensemble under consideration. Figure 6 shows that, if the data of the above instruments are used individually, then, at a 25% error of AAC measurements, an error of PSD retrieval smaller than 100% can be achieved only with the SAGE III instrument and only for particles with sizes of 0.11 to 0.61 μm (for the ensemble of realizations of stratospheric aerosol considered above). For instruments measuring the AAC in individual spectral channels of the IR region, even minimum errors in PSD retrieval are greater than 100%. Thus, the SAGE III instrument, which measures the AAC in the visible and near IR regions, is more informative with respect to PSD retrieval for submicron particles. However, if AAC measurements in all channels of the above instruments are used, an error smaller than 100% can be achieved for particles with sizes of 0.1 to 1.8 μm. Note that, in the combined interpretation of the data of the IR interferometer and SAGE III, an error smaller than 100% can be obtained for particles with sizes of 0.06 to 3.1 μm (for particles of 0.11 to 3.1 μm in a single IR experiment).

Table 3 presents the prior variability of PSD integral moments (N_{opt}, S, V) and the error in their determination from AAC measurements with errors of 5 and 25%. The results are given for different experimental schemes: for each of the SAGE III, ISAMS, CLAES, and HALOE instruments; for the four listed instruments in combination; and for the combined scheme IR (measurements in 2201 channels of the wavelength range 1.92–12.5 μm) + SAGE III. (Note that the instruments operating currently onboard the ACE satellite [35] allow the implementation of such a scheme of AAC measurements at the present time.) Retrieval errors in the case of AAC measurements for an individual IR region are presented in Table 1. From

Table 2. Spectral channels of the instruments used for aerosol monitoring

Instrument	Wavelength, μm
HALOE	2.45, 3.40, 3.45, 5.26
CLAES	3.52, 5.27, 6.23, 7.96, 10.81, 11.38, 11.86, 12.66, 12.82
ISAMS	4.54, 5.28, 6.23, 6.76, 7.40, 7.85, 8.09, 10.00, 11.36, 12.11, 16.26
SAGE-III	0.385, 0.450, 0.520, 0.600, 0.675, 0.755, 0.870, 1.020, 1.545

the data of Table 3, it follows that the total surface area and volume of aerosol particles are best determined from the AAC measurements. From comparison of individual experiments, it can be seen that S is best determined from interferometric IR measurements, where the prior uncertainty is decreased by factors of 7.6 and 6.2 at AAC measurement errors of 5 and 25%, respectively. For other instruments, the factors of a decrease in the prior uncertainty at these two AAC measurement errors are respectively 5.2 and 4 for SAGE III, 4.2 and 2.9 for ISAMS, 4.8 and 2.7 for CLAES, and 3.9 and 3.5 for HALOE. If it were possible to simultaneously use the data measured by all satellite instruments listed in Table 2, the prior uncertainty in S could be reduced by factors of 11 and 6 at 5 and 25% errors in AAC measurements, respectively. If the data of both SAGE III and the entire IR region are used, the prior uncertainty is decreased by factors of 24 and 12 at 5 and 25% errors in AAC measure-

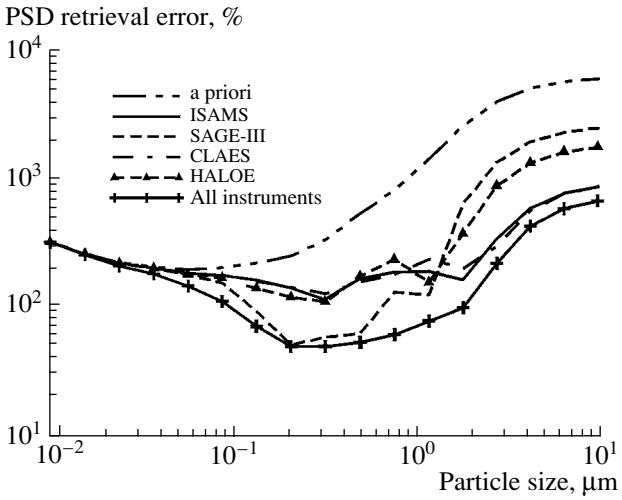

Fig. 6. Relative error in determining the PSD from the AAC values measured with an error of 25% with each of the SAGE III, ISAMS, CLAES, and HALOE instruments or with all instruments simultaneously.

Table 3. Natural variability (a priori) and absolute error in determining the integral microphysical parameters obtained by the regression method from AAC

AAC	Scheme	N_{opt}, cm^{-3}	S, $\mu m^2/cm^3$	V, $\mu m^3/cm^3$
a priori		**44.4**	**5.57**	**0.88**
5%	ISAMS	37.8	1.31	0.013
	CLAES	37.4	1.17	0.014
	HALOE	38.5	1.43	0.022
	SAGE-III	36.1	1.06	0.057
	4 instruments	**26.9**	**0.51**	**0.008**
	IR + SAGE-III	**23.4**	**0.23**	**0.003**
25%	ISAMS	40.2	1.92	0.034
	CLAES	40.2	2.03	0.043
	HALOE	38.9	1.57	0.036
	SAGE-III	38.2	1.37	0.084
	4 instruments	**32.2**	**0.91**	**0.020**
	IR + SAGE-III	**27.7**	**0.46**	**0.014**

ments, respectively. For the parameter V, AAC measurements with 5 and 25% errors make it possible to obtain the following factors of a decrease in the prior uncertainty: 15.4 and 10.6 for SAGE III, 68 and 26 for ISAMS, 63 and 20 for CLAES, and 40 and 24 for HALOE, respectively.

The combined experiment with the four instruments allows the prior uncertainty in V to be decreased by factors of 110 (5% error in AAC measurements) and 44 (25% error in AAC measurements). Comparison of the data of Tables 1 and 3 shows that the addition of SAGE III channels to IR measurements in 2201 channels does not improve the estimate for V markedly. Table 3 also indicates that the combined experiments make it possible to reduce the prior uncertainty in N_{opt} by 37–65% (four instruments) and 60–90% (IR + SAGE III) if the AAC is specified with an error of 5–25%. As for the possibility of determining other microphysical parameters, the use of both individual and combined schemes of AAC measurements with different instruments does not lead to a considerable improvement of retrieval estimates compared to the prior uncertainty for such parameters as r_g, σ, R_{eff}, and Wt. This finding indicates that the methods of solving the inverse problem of interest that are based on PSD parametrization with model (2) (see, for example, [9, 36]) cannot be efficient.

Influence of the Prior Ensemble on the Informativeness and Accuracy of the Remote Method

There is some arbitrariness in the formation of the prior ensemble of realizations of stratospheric aerosol (for example, the choice of variation ranges for the parameters of distribution (2)). On the other hand, to a certain extent, this choice determines both information characteristics of the remote method of interest and errors in retrieval of the PSD and its parameters. In particular, the relative errors in PSD retrieval obtained by us turned out to be larger than those found in a number of works devoted to analysis of the remote method under consideration [36, 37]. This circumstance is due to the fact that our ensemble of stratospheric-aerosol PSD realizations is very wide. Thus, the errors and information characteristics of the remote method under study were also calculated by us for a narrower prior ensemble.

A new ensemble (denoted by II, as distinguished from the initial ensemble denoted by I) differs from ensemble I in that, during the construction of PSD realizations in a statistical ensemble of stratospheric aerosol in accordance with the model covariance matrix (see formula (3)), we used an RMSD value of 0.35 for N at all altitudes (0.7 in ensemble I). Thus, we reduced the possible variation in the total number of aerosol particles by one-half. The other modeling parameters remained invariant. The average integral moments of the PSD were somewhat decreased because ensemble II did not contain realizations with a large number of particles in bins (the limits of parameter variations were substantially decreased from above). The variability of ensemble II as compared to ensemble I was decreased by factors of 1.6 for N_{opt}, 1.5 for S, and 1.7 for V. This implies that ensemble II is approximately 1.5 times less variable than ensemble I, while the average microphysical (and optical—AAC) parameters of ensembles I and II are rather close to one another.

Since a sufficient number of simultaneous AAC measurements with the SAGE III and HALOE satellite instruments are accumulated, we consider these ensembles precisely on the basis of retrieval of PSD parameters from AAC measurements with these instruments. AAC calculation in the indicated spectral channels for the two ensembles has shown that the ensemble-average AAC for ensemble II is 20–25% lower than for ensemble I, whereas the variability of the AAC in ensemble II is 1.5–2 times smaller than in ensemble I.

Table 4 (second and fourth rows) presents the errors in determining the PSD integral parameters (N_{opt}, S, V, R_{eff}) for the combined experiment HALOE + SAGE III with AAC measurements with a 25% error for two prior ensembles. The first and third rows of the table show the prior (natural) variability of stratospheric-aerosol ensembles I and II, respectively. The ratio of

the prior uncertainty to the retrieval error is also presented in parentheses; i.e., it is shown how many times the prior uncertainty was decreased during use of AAC measurements (informative characteristic). From comparison of the results obtained for the two ensembles, it follows that the absolute measurement errors of parameters in ensemble II are smaller than those in ensemble I. For example, the measurement errors in the PSD moments N_{opt}, S, and V are 34.5 cm^{-3}, 1.08 $\mu m^2/cm^3$, and 0.026 $\mu m^2/cm^3$, respectively, for ensemble I and 23.4 cm^{-3}, 0.77 $\mu m^2/cm^3$, and 0.020 $\mu m^2/cm^3$, respectively, for ensemble II. At the same time, owing to the wider prior variability of ensemble I, its informativeness (the ratio of the prior to the a posteriori uncertainty) for the parameters under consideration is somewhat better. For the effective radius of aerosol particles R_{eff}, narrower ensemble II yields better estimates for both the error and the informativeness.

Thus, in regard to the prior information, it can be inferred that the use of a narrower ensemble makes it possible to obtain smaller errors in retrieving microphysical parameters from AAC spectral measurements. For example, if AAC values are specified with a 25% error, the relative errors in determining S and V in the experiment HALOE + SAGE III are 42 and 14%, respectively, for wide ensemble I and 36 and 12%, respectively, for ensemble II.

5. EXAMPLES OF RETRIEVAL OF INTEGRAL PARAMETERS OF STRATOSPHERIC AEROSOL FROM AAC MEASUREMENTS WITH SAGE III AND HALOE INSTRUMENTS

Let us apply the statistical approach developed by us to analysis of satellite measurements. Consider two ensembles of AAC measurements taken with the SAGE III and HALOE instruments simultaneously in 2002–2003 and in the same regions of the Earth. The ensembles consist of 45 AAC vertical profiles in nine channels of the SAGE III instrument and four channels of the HALOE instrument (see Table 2).

Table 4. Prior uncertainty and error in determining microphysical aerosol parameters from the AAC values measured with the SAGE III and HALOE instruments with an error of 25% (the ratio of the prior uncertainty to the retrieval error is given in parentheses)

No	Scheme	N_{opt}, cm^{-3}	S, $\mu m^2/cm^3$	V, $\mu m^3/cm^3$	R_{eff}, μm^2
I	a priori	*44.5*	*5.57*	*0.883*	*0.082*
	HALOE + SAGE-III	34.5 (1.29)	1.08 (5.16)	0.026 (34.0)	0.066 (1.24)
II	a priori	*28.4*	*3.73*	*0.511*	*0.082*
	HALOE + SAGE-III	23.4 (1.21)	0.77 (4.84)	0.020 (25.6)	0.061 (1.34)

Figure 7 shows the ensemble-average vertical profiles of the total surface area and volume of aerosol particles. The profiles were retrieved from (i) AAC measurements with the SAGE III instrument, (ii) AAC measurements with the HALOE instrument, (iii) combined AAC measurements with the two instruments, and (iv) integral parameters measured by the HALOE instrument. First, Fig. 7 shows systematic distinctions between the parameters retrieved from the HALOE and SAGE-III measurements. The values of integral parameters obtained from AAC measurements with the HALOE instrument are 20–25% higher than those obtained from AAC measurements with the SAGE III instrument. In a number of studies, (see, for example, 38, 39]), similar comparisons were made for the total surface area and volume of aerosol particles obtained from SAGE II and HALOE measurements. Analysis

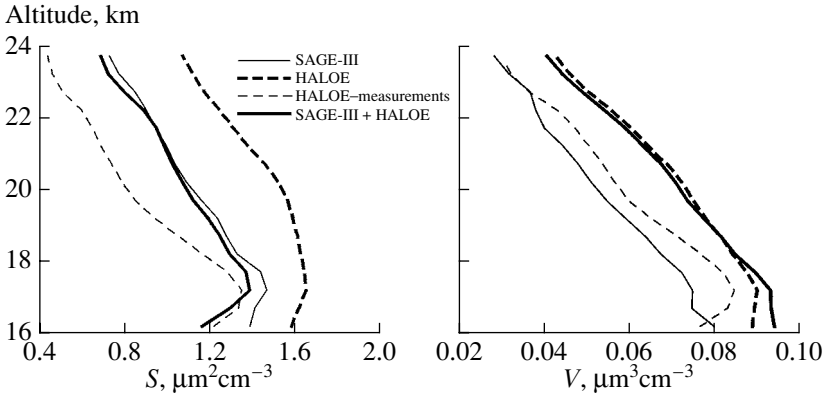

Fig. 7. Ensemble-average profiles of the total (left) surface area and (right) volume of all aerosol particles.

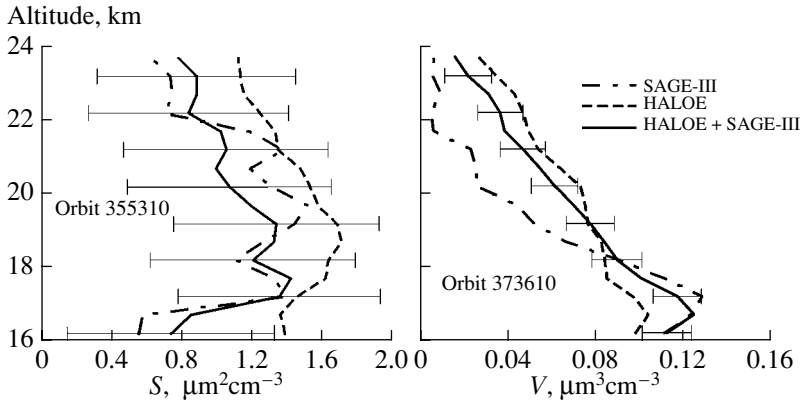

Fig. 8. Examples of retrievals of (left, orbit 355310) *S* and (right, orbit 373610) *V* from the AAC values measured with the SAGE-III and HALOE instruments.

of the results has also shown a systematic overestimation of the integral parameters retrieved from the HALOE data as compared to the SAGE II data, in particular, under the conditions of the background atmosphere at altitudes above 15 km [39].

Figure 7 also demonstrates that the retrieval of the parameters *S* and *V* from AAC measurements with the SAGE III and HALOE instruments is not a simple superposition of the results of retrieval from the data of individual instruments. First, the parameter *S* retrieved from the data of the two instruments turns out to be closer to the total surface area retrieved from SAGE III measurements alone than to that from only HALOE measurements. The opposite situation is observed for the parameter *V*. Clearly, this result is explained by both the larger contribution of large particles to the total volume in comparison with the total surface area and a higher informativeness of IR measurements with respect to large particles. Second, for both *S* and *V*, the average profiles retrieved from combined AAC measurements with the two instruments do not lie between the corresponding profiles obtained for individual instruments. The latter result points indirectly to the advantage of simultaneously using two ranges for AAC measurements. Figure 7 also indicates that the measured (contained in files with HALOE measurements) average profiles of *S* differ substantially (a difference of up to 0.9 $\mu m^2/cm^3$) from the *S* values retrieved from AAC HALOE measurements; however, these profiles are in satisfactory agreement (a difference of up to 0.3$\mu m^2/cm^3$) with the *S* values obtained from AAC SAGE III measurements and combined measurements. At the same time, for the average profiles of *V*, the measured values are in satisfactory agreement (a difference of less than 0.01 $\mu m^3/cm^3$) with the results of retrieval from AAC data obtained for different schemes.

Figure 8 illustrates individual examples of retrieval of the vertical profiles of *S* and *V* and, on the whole, confirms the inferences made from analysis of Fig. 7 that AAC SAGE III measurements make a larger contribution to the retrieval of the total surface area of aerosol particles, whereas AAC HALOE measurements contribute more to the retrieval of their total volume. Figure 8 also presents the errors of single measurements of PSD moments made on the basis of the combined scheme. It is easy to see that, for *S*, the results of all retrievals lie within the limits of this error except for the HALOE data at 16 km and, for *V*, the results of retrieval from the SAGE III and HALOE data fall within the limits of the error below and above 18 km, respectively.

6. MAIN RESULTS AND CONCLUSIONS

On the basis of analysis of a large amount of experimental data on aerosol microphysical parameters, a statistical ensemble was modeled for the background stratospheric aerosol (4800 relaizations). This ensemble includes realizations of (i) the parameters of a unimodal distribution of a log-normal PSD, (ii) the aerosol composition (percentage of sulfuric acid aerosol), and (iii) the temperature and humidity at altitudes of 15 to 30 km. The following results were obtained with the use of this ensemble.

(1) Algorithms of the Mie theory were used to calculate the aerosol attenuation coefficients (AACs) in the wavelength range 0.38–16.3 μm for all ensemble realizations. Analysis of the correlation coefficients (CCs) between the AAC and different aerosol characteristics has shown that the AAC correlates most strongly with the total volume *V* (CC = 0.9–1.0) and surface area *S* (CC = 0.6–0.8) of all particles.

(2) The errors in determining microphysical parameters from AAC measurements in the IR region of the spectrum (1.92–12.5 μm) were analyzed via the method of multiple linear regression. It has been shown that, at a 5% error of AAC measurements, the prior PSD uncertainty for particles with sizes of 0.4 to

4 μm is decreased by more than an order of magnitude (from 500–5000% to 20–50%). The errors in S and V retrievals are 7.6 and 293 times smaller than the prior uncertainty (29 and 1.5% of the ensemble average), respectively. The error in determining such parameters as r_g, σ, N_{opt}, R_{eff}, and Wt are decreased only slightly as compared to the prior uncertainty (by 5–30%).

(3) Spectral schemes of AAC measurements with the SAGE III, ISAMS, CLAES, and HALOE instruments and an IR interferometer, as well as combined measurement schemes for determining microphysical characteristics of the background stratospheric aerosol, were discussed. It has been shown that the use of the combined schemes makes it possible to extend the range of particle sizes yielding a satisfactory accuracy of PSD retrieval and to considerably reduce the error in determining such PSD integral parameters as S and V and, to a smaller extent, the total number of particles N_{opt}.

(4) The influence of the prior statistics (ensemble width) on the informativeness and errors of the remote method of interest was analyzed. It has been shown that the use of a narrower ensemble makes it possible to obtain smaller errors in retrieving microphysical parameters from AAC spectral measurements.

(5) Examples of interpretation of the data of AAC measurements with the SAGE III and HALOE instruments are presented for the cases of their spatial and temporal coincidence. A systematic disagreement of the vertical profiles of S and V obtained from SAGE III and HALOE measurements was revealed. This finding calls for further studies to provide possibilities of forming a unified climatological data bank on aerosol parameters of the background stratosphere.

ACKNOWLEDGMENTS

This study was supported by the Russian Foundation for Basic Research (project nos. 03-05-64626, 05-05-65305), the Russian Universities grant (no. UR.01.01.044, and the NASA NAG (grant no. 5-11248).

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