Optimal Eigenanalysis for the Treatment of Aerosols in the Retrieval of Atmospheric Composition from Transmission Measurements.

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Abstract

The separation of the individual contributions of aerosols and gases to the total attenuation of radiation through the atmosphere has been the subject of much scientific investigation since remote sensing experiments first began. Here we describe a new scheme to account for the spectral variation of the aerosol extinction in the inversion of

transmission data from occultation measurements. Because the spectral variation of the aerosol extinction is generally unknown, the inversion problem is underdetermined and cannot be solved without reducing the number of unknowns in the set of equations used to describe the attenuation at each wavelength. This reduction can be accomplished by a variety of methods including the use of *a priori* information, the parameterization of the aerosol spectral attenuation, and the specification of the form of the aerosol size distribution. We have developed and implemented a parameterization scheme based upon existing empirical and modeled information about the microphysical properties of aerosols. This scheme employs the eigenvectors from a very extensive set of simulations to parameterize the aerosol extinction coefficient for incorporation into the inversion algorithm. We examine the accuracy of our method using data sets containing over 24,000 extinction spectra and compare it with that of another scheme which is currently implemented in the POAM (Polar Ozone and Aerosol Measurement) satellite experiment. In simulations using eighty wavelengths in the UV-vis-near IR spectral range of the SAGE (Stratospheric Aerosol and Gas Experiment) III instrument we show that, for our optimal parameterization, errors below 1 % are observed in 80 % of cases, whereas only about 20 % of all cases are as accurate as this in a quadratic parameterization employing the logarithm of the wavelength.

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Introduction

The atmospheric aerosol has a pronounced effect on radiative transfer in a wide spectral region from the ultraviolet through the infrared[1]. It is necessary therefore, to take the effect of aerosols into account in the solution of direct and inverse problems in atmospheric optics[2,3]. Such problems include the determination of the concentration of trace gases from transmittance measurements, the differentiation between aerosols and molecular continua in remote measurements in the thermal spectral region, and the determination of aerosol physical properties from transmission and scattering measurements. To solve such problems one has to employ simplifying assumptions or *a priori* information about the aerosols. In this work we shall address one of these problems in particular—that of determining the composition of the atmosphere from satellite measurements of atmospheric transmittance, where there is a direct relationship between the exact determination of the atmospheric gaseous content and the correct description of the spectral variation of aerosol extinction. Here, the separation of the effects of aerosols and gases involves the specification of the spectral behavior of the aerosol extinction coefficient in one form or another (either indirectly or explicitly).

A number of works demonstrate the importance of solving this problem correctly[4-7]. Steele and Turco[4] showed that the operational algorithm used to process the SAGE II measurements introduced systematic errors in the retrieval of the ozone concentration due to an incorrect approximation of the spectral behavior of aerosol extinction. This error could be as much as 15 % in the ozone content of the lower stratosphere. Application of an alternative algorithm made it possible to bring about agreement between the SAGE II ozone trends in the lower stratosphere and those of independent measurements (ozonesondes and Umkehr).

Rusch et al.[5] employed a different algorithm to account for the aerosol contribution in the SAGE II measurements. They showed that for altitudes above 22 km, where the aerosols account

for a relatively small proportion of the total extinction in the ozone channel, the operational and their new algorithm produce agreement in ozone concentration to within 1 %, but below 22 km, where the aerosol contribution is larger, their algorithm yielded an ozone content considerably lower (by 30 %) than the values obtained by the operational algorithm, and were in better agreement with ozonesonde data.

In the retrieval of gas and aerosol concentrations from transmission measurements, the gas spectral attenuation is uniquely determined by its spectroscopic characteristics, which depend only on temperature and pressure, while the spectral behavior of the aerosol extinction is governed by a large number of parameters. These parameters include total concentration, the size distribution of the aerosol components, particle shape and structure, and the complex refractive index. Many of these aerosol characteristics are related to the ambient conditions such as temperature and H_2O partial pressure. In the simulations presented here solar occultation viewing geometry is applied to the problem of retrieving aerosol extinction in the upper troposphere and lower stratosphere. We apply *a priori* information about the environmental conditions and the physical properties of the aerosols in this region of the atmosphere in our retrieval. But, because the spectral behavior of the aerosol extinction is unknown, regardless of the number of instrument channels the system of equations will be underdetermined. This results from the fact that the number of variables to be retrieved from measurements at n wavelengths is equal to total number of optically active gases (N_g) plus the aerosol extinction at each wavelength (n values). Therefore in order to solve the inverse problem some assumptions must be made about the spectral correlations of the aerosol optical properties in the different channels.

Statement of the problem

The transmittance method is based on the law of attenuation of solar, lunar and stellar radiation through the atmosphere (the Bouguer–Beer–Lambert law) along a tangential path as the radiation source rises or sets on the planet's horizon. The attenuation of radiation of wavelength λ propagating through the atmosphere along a trajectory S is characterized by the transmittance, $P(\lambda,S)$:

$$P(\lambda,S)=\frac{I(\lambda,S)}{I_0(\lambda)} \tag{1}$$

where $I(\lambda,S)$ is the intensity of radiation which passes through the atmosphere and $I_0(\lambda)$ is the intensity of the radiation above the atmosphere. At temperature *T(z)* and pressure *p(z),* contributions to the atmospheric optical depth come from gas absorption—a product of the gas density, $\rho_i(z)$, and the absorption coefficient, $K_i\ (\lambda,T(z),p(z))$ — Rayleigh scattering, $\beta_R(\lambda,z)$, and aerosols, $\beta_a(\lambda,z)$. If N_g is the number of optically active gases, *z* is the height, and *s* is an element along the path, *S*, we consider the problem of determining the atmospheric gas and aerosol profiles, $\rho_i(z)$ and $\beta_a(\lambda,z)$ from the transmittance, *P(λ,S)*:

$$P(\lambda,S)=\exp\left(-\int_S\left(\sum_{i=1,N_g}\rho_i(z(s))K_i(\lambda,T(z(s)),p(z(s)))+\beta_a(\lambda,z(s))+\beta_R(\lambda,z(s))\right)ds\right) \tag{2}$$

The approximation of a spherically stratified atmosphere is convenient for representing the aerosol extinction coefficient, $\beta_a(\lambda,z)$, in the form of parametric functions $\beta_a^z(\lambda)$, which characterize the spectral dependence of the aerosol extinction at the height z. For homogeneous

spherical particles these functions are given by a physical (Mie) theory describing radiation attenuation by aerosols through the relationship:

$$\beta_a^z(\lambda) = \sum_j \int_0^\infty \pi r^2 Q_j(m_j, r, \lambda) f_j(r) dr \tag{3}$$

Here, j is the index denoting an individual aerosol type, r is the particle radius, $f_j(r)$ is the size distribution function, and $Q_j(m_j, r, \lambda)$ is the Mie extinction coefficient of a particle with refractive index m_j. The number of particles of type j per unit volume of the atmosphere is given by $N = \int_0^\infty f_j(r) dr$. Equation (3) can be used as a basis for deriving a simple relationship between the aerosol extinction at different wavelengths, and as such, is employed in many of the schemes used to solve the inversion problem.

Review of available methods

There are many schemes available for parameterizing the spectral dependence of aerosol extinction[7-15]. The fewer the number of parameters employed, the greater the *a priori* constraint on the retrieval. When multi-wavelength data exist it is common to employ a number of free parameters, which can have physical meaning in the aerosol size distribution model, or be simply used to model the spectral shape of the extinction data. One of the most common methods employed for stratospheric aerosols has been the use of a uni-modal lognormal size distribution function, given by:

$$f(r) = N \frac{\exp\left\{-\ln^2\left(r / r_g\right) / \left(2 \ln^2 \sigma\right)\right\}}{r\sqrt{2\pi}\ln\sigma} \tag{4}$$

where r_g is the median (also the geometric mean) radius, σ, the geometric standard deviation, and $N = \int_0^\infty f(r)dr$, is the total number of particles per unit volume of air. This model has been used in schemes with one free parameter, N (Hervig et al.[7]), with two free parameters, N and r_g (Hervig et al.[12]) and with three, N, r_g and σ (Rusch et al.[5]), and applied to aerosol retrieval from the HALOE (Halogen Occultation Experiment) and SAGE measurements. One of the problems encountered in employing a scheme of this type is the non-linearity of the inversion, which makes the method computationally slow and sometimes difficult to achieve convergence and reach a solution.

POAM uses a three-parameter aerosol retrieval scheme based on a Taylor expansion of the second degree (Lumpe et al.[13]). The extinction is modeled by the expression:

$$\ln \beta_a(\lambda, z) = \mu_0(z) + \mu_1(z) \ln \lambda + \mu_2(z)(\ln \lambda)^2 \tag{5}$$

where λ is the wavelength and the μ_i are free parameters. Since the spectral variation of the extinction tends to be quite smooth in the UV-vis-near IR region of the spectrum, this gives an adequate representation of it in most cases. However, there is no physical basis for the expression, and the free parameters have no physical significance. Again, non-linearity can present a problem in the inversion.

The SAGE II operational scheme (Chu et al.[14]) is based on a linear constrained solution to (3), in which a linear relationship between the $\beta_a^z(\lambda)$ at different wavelengths was developed to formulate an expression relating the aerosol extinctions at five of the wavelengths:

$$\beta_a(\lambda_5) = \sum_{i=1,4} \alpha_i \beta_a(\lambda_i), \tag{6}$$

where λ_i, $i = 1,...,5$ are the wavelengths of the measurement channels. The coefficients α_i, $i = 1,..,4$ are calculated according to the Mie theory, and the Tikhonov regularization method is used to solve the ill–posed problem. Steele and Turco[4] analysed the eigenvalues of the Mie extinction cross-section to derive these coefficients. These schemes impose a linear relationship between the aerosol extinctions and hence decrease the number of independent parameters by 1. The advantages of such methods are that the coefficients themselves are based on the physics of the scattering problem, and the inversion scheme is linear. However, the solution will depend on the coefficients chosen, which do not necessarily provide the optimum solution under both quiescent and volcanic conditions (Steele and Turco[4]).

The accuracies of the various schemes are different for every atmospheric situation and depend not only on the aerosol content, but also on the constituent gases. Chu et al.[14] provide estimates of the errors in the operational algorithm used to process the SAGE II measurements and Thomason[16] showed that the application of the Angstrom law to SAGE II aerosol extinction data could result in errors of 5–10 % in the extinction coefficient. According to the estimates given by Fussen[6] the operational SAGE II algorithm yields a 5–15% error in aerosol extinction at a wavelength of 0.6 μm. He showed how a cubic fit to the aerosol extinction spectral dependence could give noticeably better results. Steele and Turco[4] estimated the r.m.s. (root mean squared) error of the SAGE II algorithm to be 7 % at 0.6 μm wavelength, with maximum errors reaching 29 %. In simulations at the POAM II wavelengths, Lumpe et al.[13] estimated random errors of 10-20% in the retrieved 1.06 μm extinction using the quadratic parameterization.

It would be logical to expect that the greater the number of parameters used in representing the aerosol extinction spectral dependence, the greater the accuracy of that

approximation. However, an increase in the number of free parameters causes a relaxed conditionality of the corresponding equations, which is not necessarily consistent with the information content of the transmittance measurements. The equations should be satisfied only to within the error of the measurements, as exact agreement can force unphysical solutions in the underlying aerosol properties[17,18]. Therefore the accuracy of parameterizing the spectral behavior of aerosol extinction should be achieved not merely by increasing the number of free parameters but also by employing *a priori* knowledge to select an optimal set of basis vectors in which to express the spectral variation of the aerosol extinction.

An optimal method of parameterization is characterized by certain features. First, adequate and general *a priori* information on the microphysical (or optical) characteristics of the atmospheric aerosol should be used. From this standpoint, the use of a single size distribution function, *f(r),* to generate the aerosol extinction spectral dependence is inadequate. Second, the number of free parameters must be optimal. In this case, it is useful to employ the ideas of optimal parametrization given by Oboukhov[19], which were first used by Biryulina and Rozanov[15] for parameterizing the aerosol extinction. Third, in solving the inverse problem, it is best to employ the maximum amount of data (traditional or satellite) to specify the state of the atmosphere during measurements. Instruments with enough measurement channels can supply additional information that can be used in the inversion. For example, data on water vapor concentration and temperature, and on the concentration of SO_2 and other gases that contribute to aerosol formation can be used to characterize the aerosol, and thus constrain the solution to the inverse problem. In addition to achieving the most accurate representation of the spectral variation of the aerosol extinction, for computational efficiency and stability it is preferable to employ a linear algorithm.

Description of proposed algorithm

The algorithm developed here uses existing empirical and modeled information on the microphysical properties of the aerosol (composition, size distribution) to generate a set of aerosol extinction spectra. Our calculations employ Mie theory, which is based upon single-scattering from spherical particles. Although some atmospheric particles may not be spherical, nearly all those encountered in the stratosphere are expected to be, with the possible exception of uncoated dust particles and frozen particles found in cirrus clouds and some polar stratospheric clouds, which will be treated in future work. The first few eigenvectors of the covariance matrix based on these spectra are the pseudoempirical orthogonal functions which can be used to optimally model the spectral behavior of the aerosol extinction.

Aerosol models

In order to obtain a representative (global) data set on stratospheric aerosol optical spectra for use in forming the basis set, a statistical simulation of the spectral and height dependence of the stratospheric aerosol was carried out[20]. Data on the structure, dispersivity, and chemical composition of stratospheric aerosols were compiled, and the variance of these quantities estimated from published data. Using these statistics, random realizations of the microphysical model were generated, and from these, aerosol extinction spectra as a function of altitude.

The composition and size distributions of the aerosols included in the data set were based on the published data of Krekov and Zvenigorodski[21], Lazrus and Gandrud[22], Pinnick et al.[23], Toon and Pollack[24], Rosen et al.[25], Gras and Michael[26], and Turco et al.[27]. These include sulfuric acid aerosol drops resulting from chemical and photochemical reactions between SO_2, H_2O, and

O_2; volcanic dust and meteoritic dust. The meteoritic dust is the product of meteoritic combustion and, according to Krekov and Zvenigorodski[21], can be divided into two fractions—fine–dispersed (referred to as "meteoric–1 dust") and coarse–dispersed ("meteoric–2 dust"). The modeled particles also include two–layer aerosols with cores of volcanic and meteoric–1 dust and an outer layer of sulfuric acid solution. Through heterogeneous nucleation these particles can be large with a complex and variable chemical composition[22,28] including H_2SO_4.

The refractive indices for both volcanic and meteoritic dust are taken from Krekov and Zvenigorodski[21], and their weight variations estimated on the basis of the same data. In order to account for the refractive index data of different authors[21,28-32] weighted means are employed where the weights, p_i, are the fraction of total mass of each aerosol constituent:

$$m(\lambda) = \sum_{i=1}^{N} p_i m_i(\lambda) \Big/ \sum_{i=1}^{N} p_i \ . \tag{7}$$

Here λ is the wavelength, and $m_i(\lambda)$ the refractive index of each individual component. N is the number of components. The composition of the sulfuric acid aerosol is determined by the temperature and concentration of water vapor according to Steele and Hamill[33] and the refractive index taken from Palmer and Williams[30]. Thus, the variations in the refractive index of the sulfuric acid aerosols are governed by the variations in temperature and water vapor profiles.

The size distribution of each type of aerosol is based upon a generalized lognormal distribution[21,27] with three varied parameters r_g, σ and b:

$$f(r) = \frac{N r^b}{r_g^{b+1} \ln\sigma\sqrt{2\pi}\exp\left(\frac{\ln^2\sigma}{2}(b+1)^2\right)} \exp\left(-\frac{\ln^2(r/r_g)}{2\ln^2\sigma}\right). \tag{8}$$

The means of the distribution parameters and their variations are taken from Krekov and Zvenigorodski[21]. N is the total number of particles per unit volume of air. Using equation (8) with the parameter b equal to –1 reduces the generalized lognormal size distribution to the unimodal lognormal size distribution (eq. 4). Two models were used to generate the value of the parameter b, based on the data of Krekov and Zvenigorodski[21]—the base model used a value of $b = -3$ with a standard deviation of 0.1, and the second model used a value of $b = -2$ with a standard deviation of 0.8. The vertical profiles of the aerosol concentrations and the parameters of the distribution function are simulated as normally distributed random vectors[34] characterized by a certain mathematical expectation and a covariance matrix for which the following model is used[35]:

$$\mathrm{cov}(X)(z_i, z_j) = \sigma_X(z_i)\sigma_X(z_j)\exp(-|\, z_i - z_j \,|/r_C), \tag{9}$$

where $\sigma_X(z_i)$ and $\sigma_X(z_j)$ are the standard deviations in the parameter X at the heights z_i and z_j, and r_C is the correlation radius. Typical values for these for a number of different parameters are given in Table 1. In our statistical simulations the total number of varying parameters is about 30.

In modeling the vertical variation of the different parameters, we take into account the cross correlations between them. For this purpose, the total vector of the parameters is simulated on the basis of a total covariance matrix. This matrix has a block structure containing both auto-covariance (9) and cross–covariance (between different parameters) matrices as blocks. The cross–covariance matrices are constructed on the basis of the following considerations. It is assumed that the atmospheric aerosol layers correspond to temperature inversion regions[21]. This condition is represented in our model by a statistically strong positive correlation between the

temperature gradient and the concentrations of sulfuric–acid, volcanic, and meteoritic aerosols. A correlation coefficient of 0.8 is employed for all heights and types of aerosols. For the size parameters related to the sulfuric–acid aerosols, an obvious correlation with the concentration of sulfur dioxide (the basic source of these aerosols) is included in the model, such that the distribution function parameters r_g and σ increase with an increase in the SO_2 concentration. Therefore we include correlation coefficients of 0.7 (the quantitative expression of the notion of "strong correlation") between the SO_2 and sulfuric–acid aerosol concentration and the parameters r_g and σ, for all heights.

As noted above, two–layer particles with shells of sulfuric acid are a constituent part of the model. The contribution of heterogeneous nucleation to shell formation is still little understood[21]. Therefore the portion of the indicated particles is simulated as a random number to take into account the full range of possible variations. The model of shell growth is that of Vasilyev and Ivlev[37], corrected so that the H_2SO_4 mass is proportional to the SO_2 concentration. Hence, the shell radius is proportional to the cubic root of the SO_2 concentration. With this correction the model of shell growth can be expressed as:

$$r / r_c = 1 + CQ^{1/3}, \tag{10}$$

where r is the total particle radius, r_c is the core radius, Q is the SO_2 concentration in cm^{-3}, and C is an empirical constant. The value of C is allowed to vary randomly based on the estimates of Vasilyev and Ivlev[37] with a mean of 0.001 and a standard deviation of 100%. The model includes a parameter for the core solution in the shell, which varies according to the core type. For describing the core solution, a coefficient which varies between 0 (for an insoluble core) to 1 (for complete dissolution) is introduced. For quartz the value is 0, for ammonium sulfate it

depends upon the concentration of the sulfuric acid solution, and for dust it is close to 0. The solubility data used are taken from Goronovski at al.[38].

Determination of the orthogonal basis set

Calculations of the aerosol extinction coefficients were performed using Mie theory and a modified algorithm for ensembles of homogeneous particles[39,40], and special algorithms for ensembles of two-layer particles[37,41]. Five hundred atmospheric situations (s = 1, 500) were chosen and for each, the set of aerosol extinction coefficients $\beta_s(\lambda_i, z_j)$, was calculated at 49 altitudes, $z_j = 11 + j$ km (j = 1, 49), for λ_i from 250 to 1560 nm. Eigenvectors were determined for extinction in the SAGE III (http://www-sage3.larc.nasa.gov/) spectral region—0.29 to 1.03 μm plus 1.52 to 1.56 μm at a total of 80 wavelengths. Thus, the total number of simulations of aerosol extinction spectra used for calculating the eigenvectors was 24,500 (49 x 500). This is referred to below as the main data set.

From these extinction spectra the mean vectors for both the extinction and its logarithm were derived:

$$\overline{\widetilde{\beta}}(\lambda_i) = E\left(\widetilde{\beta}(\lambda_i)\right) \approx \frac{1}{500}\frac{1}{49}\sum_{\substack{s=1,500\\ j=1,49}} \widetilde{\beta}_{s,j}(\lambda_i) \text{ and} \tag{11}$$

$$\overline{\ln \widetilde{\beta}}(\lambda_i) = E\left(\ln \widetilde{\beta}(\lambda_i)\right) \approx \frac{1}{500}\frac{1}{49}\sum_{\substack{s=1,500\\ j=1,49}} \ln \widetilde{\beta}_{s,j}(\lambda_i) \tag{12}$$

The covariances $K_\beta = \{k_{\beta i,j}\}_{\substack{i=1,n\\ j=1,n}}$, ($n$ = 80, the number of wavelengths) were then calculated according to:

$$k_{\beta i,j} \approx \frac{1}{500 \times 49 - 1} \sum_{\substack{s=1,500 \\ r=1,49}} \left(\widetilde{\beta}_{s,r}(\lambda_i) - \overline{\widetilde{\beta}}(\lambda_i) \right) \left(\widetilde{\beta}_{s,r}(\lambda_j) - \overline{\widetilde{\beta}}(\lambda_j) \right) \tag{13}$$

in similar way:

$$k^L{}_{\beta i,j} \approx \frac{1}{500 \times 49 - 1} \sum_{\substack{s=1,500 \\ r=1,49}} \left(\ln \widetilde{\beta}_{s,r}(\lambda_i) - \overline{\ln}\, \widetilde{\beta}(\lambda_i) \right) \left(\ln \widetilde{\beta}_{s,r}(\lambda_j) - \overline{\ln}\, \widetilde{\beta}(\lambda_j) \right) \tag{14}$$

The covariance matrices, K_β and K_β^L, can be used as *a priori* information for solving the complex inverse problem of retrieving the gas vertical profiles and aerosol extinction spectra by statistical regularization[42,43] (i.e. optimal estimation). In this paper, however, they are used in another way. It is well known that the first few eigenvectors, $f_p(\lambda_i)$, of these covariance matrices can be used to obtain the optimal approximation of the functions $\widetilde{\beta}_{s,j}(\lambda_i)$ through expansion[19,44].

$$\widetilde{\beta}(\lambda_i) \approx \overline{\widetilde{\beta}}(\lambda_i) + \sum_{p=1,n} a_p f_p(\lambda_i), \text{ and} \tag{15}$$

$$\ln \widetilde{\beta}(\lambda_i) \approx \overline{\ln}\, \widetilde{\beta}(\lambda_i) + \sum_{p=1,n} a^L{}_p f^L{}_p(\lambda_i) \tag{16}$$

The coefficients a_p and a_p^L depend on the functions $\widetilde{\beta}(\lambda_i)$ and can be calculated from the relationships:

$$a_p = \sum_{i=1}^{80} \widetilde{\beta}(\lambda_i) f_p(\lambda_i) \tag{17}$$

$$a^L{}_p = \sum_{i=1}^{80} \ln \widetilde{\beta}(\lambda_i) f^L{}_p(\lambda_i) \tag{18}$$

Eigenanalysis

The first few eigenvalues of the spectral covariance matrices K_β and K_β^L are presented in Table 2. These allow one to estimate the approximation quality as follows. The variable d_m, defined as:

$$d_m = \frac{\sum_{\alpha=1,m} e_\alpha}{\sum_{\alpha=1,n} e_\alpha},$$

where e_i is the i^{th} eigenvalue (in order of decreasing value), is commonly used in determining the error of the approximation due to terminating the expansion after the first m terms. This ratio characterizes the part of the total dispersion accounted for by the first m natural components. The values of d_m for the matrices K_β and K_β^L are given in Table 2 and demonstrate that the total dispersion is almost totally accounted for by using the first two or three vectors for both types of parameterization. But this fact does not mean that it will suffice to set bounds to 2 or 3 expansion terms in (15) and (16). The decision criterion for the number of expansion terms is based upon the required approximation errors – absolute errors in the case of a linear approximation and relative errors in the case of a logarithmic approximation.

The first few basis functions $\{f_p{}^L(\lambda_i)\}$ derived from the logarithm of the aerosol extinction coefficient are shown in Figure 1. It can be seen that the spectral dependence of the eigenvectors becomes more complex as their number increases (i.e. as their associated eigenvalue decreases). Such behavior is rather typical. For most natural functions, the high-frequency variability components are associated with small eigenvalues.

The influence of the number of basis functions employed (the free parameters) on the errors of the approximated aerosol extinction spectra through optimal parameterization are shown in the examples of Figures 2a and 2b. These show the spectral dependence of the relative errors (i.e. the difference between the actual and the approximated aerosol extinction divided by the actual extinction) when different numbers of eigenvectors are used in the approximation. When only the first eigenvector is used the approximation describes only the main overall value, but cannot describe the shape of the spectral dependence (see Figure 1, which shows the first vector to be nearly constant over the spectrum). As a result, in the examples given in Figures 2a and 2b, the approximation errors grow large at both ends of the spectrum and exceed 60 % and 120 % (not shown in Figure 2b) in the vicinity of 1.5 μm. A second eigenvector enables us to take into account the specific slope of the extinction spectral dependence and hence to describe it (to a first approximation) for different example cases. With this addition the maximum errors are reduced to less than 10 and 20 % in the examples shown in Figures 2a and 2b. To consider non–linearities in the spectral dependence it is necessary to invoke the subsequent vectors (see Figure 1). For the cases shown in Figures 2a and 2b, three and four vectors (respectively) must be used to decrease the average approximation error over the spectrum to less than 1 %.

Although Figure 2 provides an insight into the accuracy of the approximation for individual aerosol cases, it does not adequately represent the approximation errors for all possible aerosol spectra. In Figure 3 the spectral dependence of the r.m.s. relative error is shown for the entire main data set according to the number of eigenvectors employed. One can see from this how the error decreases with an increasing number of basis functions employed. When only the first eigenvector is used, the r.m.s. relative error reaches as much as 40% over the spectrum. If a second vector is added, the maximum error in the spectrum is reduced to 10%.

With a third, it is reduced to 3%, and, even though the error still varies over the spectrum, its variations are smaller than those for one or two vectors. When four or five vectors are used, the error continues to decrease, but its spectral behavior does not change radically. Initially, an increase in the number of vectors employed results in a large reduction in the aerosol approximation error, but once three or four basis vectors are employed the gain in accuracy diminishes as the number increases.

Table 3 shows the values of the relative r.m.s. error, averaged over the spectrum, for different sets of aerosol data and both linear and logarithmic parameterizations as a function of the number of basis vectors employed. The approximation errors are given as absolute and relative values for the linear parameterization and as relative values (percents) for the logarithmic one. The middle column of parts a), b) and c) shows data for the main data set described above. The next column of each part shows errors for an independent data set described below. The number of parameters needed to achieve a specified r.m.s. error in the approximation can be gauged readily from these data. For the linear parameterization, the use of 3, 4 and 5 vectors give accuracies in approximating the aerosol extinction of 0.615E-5, 0.223E-5 and 0.136E-5 km^{-1} respectively, equivalent to relative errors of 10.2, 2.3 and 1.3 %. For the same simulations the accuracies of the logarithmic approximations are between 24.5 and 2.0 %.

By employing more eigenvectors we can increase the accuracy of the parameterization but the cost of this is an increase in the propagation of measurement (random) error[4]. Thus there is a trade-off between the systematic error caused by a poor approximation and the random error caused by amplification of the experimental error by employment of the eigenvectors corresponding to the smaller eigenvalues. From Table 3 it would appear that four eigenvectors

can quite adequately represent the spectral variation of the extinction for a wide range of modeled stratospheric aerosol conditions.

Independent data sets

Table 3 and Figure 3 show the estimates of the approximation errors for the set of stratospheric aerosol states used to develop the pseudoempirical eigenvectors (i.e. the main data set). This demonstrates the accuracy of the optimal parameterization scheme but not its applicability to other stratospheric states. We therefore also tested its accuracy using independent data not employed in constructing the basis vectors.

Unfortunately, we have insufficient in situ data on the spectral dependence of the aerosol extinction to give a good estimate of the parameterization errors from these alone. Therefore, we have constructed another (independent) set of aerosol simulations to generate a statistically independent ensemble. This independent data set was created from the size distribution data of Stevermer et al.[45] and Anderson et al.[46]. In the first of these papers, parameters are listed for uni- and bimodal lognormal size distributions of aerosols for 134 cases representing both background and volcanic states compiled from different publications. The parameters of 117 of these distributions, which can be found at http://www.srrb.noaa.gov/research/aerosol.html, were obtained by direct measurements fitted to lognormal functions. The aerosol extinction was computed for these experimental size distributions assuming the particles to be spherical, homogeneous, and composed of an aqueous solution of H_2SO_4 of constant concentration.

Figure 4 shows the calculated mean extinction spectra and its dispersion for the main data set (used in the computation of the eigenvectors) and the independent data set. It can be seen that the independent set differs essentially from the main data set in the absolute values of the aerosol extinction coefficient, its dispersion, and even slightly in its spectral behavior. The mean

extinction spectrum of the independent data set lies above that of the main data set by about two orders of magnitude. This large difference is the result of two factors. First, the main data set is representative of the background stratosphere and hence corresponds to a relatively low stratospheric turbidity, whereas the independent data set represents a post–eruption highly turbid stratosphere. Second, the independent data are from the lower stratosphere where the aerosol extinction is greatest, whereas the main data set covers the height range up to 60 km and hence includes a large number of cases of very low aerosol concentration. Since the independent data set is vastly different from the simulated one it provides an excellent test for the validity of the proposed parameterization algorithm under the wide range of aerosol conditions that might be encountered in the real stratosphere.

The last column of each section in Table 3 gives the r.m.s. error in the aerosol extinction approximation for the independent data set over the spectral range considered. It can be seen that the relative errors for the independent data set are small (less than 10%) when the number of parameters is 3 or more. In a number of cases the independent set error is less than the main one used to construct the basis set. These approximation errors demonstrate that the optimal parameterization can be employed for a wide variety of aerosol cases, and that if at least four eigenvectors are employed the r.m.s. error will likely be less than 4 %.

Comparison with independent parameterization method

It is interesting to compare the performance of the optimal parameterization method with that of a popular alternative algorithm employed in the POAM satellite experiments[13,47]. The expression for the spectral variation of the aerosol extinction is given by a quadratic in the logarithm of the wavelength (equation 5). In our approach, an orthonormal basis of the eigenvectors of the covariance matrix is used. This provides an optimal approximation of the

functions under consideration. The quadratic parameterization is an expansion in terms of non-orthogonal functions $(\ln\lambda)^p$, $p = 0,1,2$ and does not have a physical basis, but simply provides a model for a smooth spectral dependence.

To analyze the accuracy of the quadratic parameterization, the calculations were made for the same sets of simulated aerosol extinction data described above. Table 4 shows a comparison of the distribution of errors for the optimal linear approximation and the quadratic expansion. The total number of cases simulated is that of the main data set of 24,500 spectra. These data show that for the optimal parameterization, errors below 1% are observed in 80 % of the cases. For the quadratic parameterization only about 20 % of all simulations are as accurate as this. Similarly, errors below 2 % are observed for the optimal parameterization in 96 % of the cases, and, for the quadratic parameterization – in only 40 % of all cases. Table 4 also shows the behavior of the parameterizations in more problematic cases, when the properties of the function do not allow it to be approximated so well. In these cases, the optimal parameterization has advantages as well. Errors over 5 % are observed in less than 0.1 % of cases in which the optimal scheme was used, whereas, for the quadratic parameterization, such cases comprise 16 %.

Figure 5 shows the spectral dependence of the aerosol extinction, $\widetilde{\beta}_{s,j}(\lambda_i)$, for the 3 worst cases (those of greatest error) for each of the parameterizations. These cases are difficult to model due to relatively abrupt changes in the slope of the spectrum. Such behavior cannot be well-described by a second–degree polynomial. In the worst cases the errors in the quadratic parameterization are almost four times those in the optimal parameterization. Figure 6 presents one of the cases of maximal error (5.8 %) for the optimal parameterization. The complexity of the approximation is caused by a noticeable inflection of the extinction curve in the vicinity of

600 - 700 nm. Figure 7 demonstrates a case of one of the largest errors (r.m.s. error of 19 %) for the quadratic parameterization and again demonstrates the difficulty in matching the spectrum when the slope changes abruptly.

By including representative spectra in the set used to construct the eigenvectors, the optimal parameterization method has the potential to accurately model non-smooth spectra, whereas the quadratic parameterization can only model smooth spectra with a single maximum or minimum.

Summary

In the problem of retrieving the characteristics of the atmospheric composition from slant path transmittance (occultation) measurements, the aerosol effect is taken into account by specifying (implicitly or explicitly) the spectral dependence of the aerosol extinction. Analyses of the results of the SAGE II and HALOE atmospheric transmittance measurements by various researchers have shown that the spectral parameterization type influences to a great extent, the solution of the inverse problem, for example, in the problem of retrieving the vertical profiles of the ozone content in the lower stratosphere. In the work described here, an optimal parameterization method is developed based upon a set of simulated aerosol extinction spectra. A pseudoempirical orthogonal basis set is defined from an ensemble of stratospheric aerosol models, constructed by random simulation using current ideas on the microphysical characteristics of stratospheric aerosol and its variability. To generate these simulated data, thirty parameters, like the humidity, temperature and SO_2 content of the atmosphere, which influence the structure and composition of the aerosol were varied over a wide range. The spectral coefficients of aerosol extinction were calculated for the SAGE III spectral region, and from these, the eigenvectors and eigenvalues were calculated. These eigenvectors were then

applied to parameterize the aerosol extinction spectral dependence. We have shown that the scheme developed here allows one to approximate the aerosol extinction spectrum for a wide variety of stratospheric aerosols with a mean relative error of 2.1 – 3.2 % for different types of optimal parameterization (linear and logarithmic) for two different data sets when four independent parameters are used, and with a mean relative error of 6.3 – 24.5 % when three independent parameters are used. The quadratic parameterization used in the POAM retrieval algorithm[13] and expressed in equation (5) uses 3 parameters and produces approximation errors in the aerosol extinction coefficients which are almost twice those of the optimal schemes developed here.

The advantages of the optimal method over the quadratic parameterization include its potential to model a non-smooth spectral variation in the aerosol extinction, its employment of *a priori* information on stratospheric aerosols, its linearity—which aids the speed and convergence of the mathematical inversion process, and its basis in physical principles. In a companion paper extensive simulations of the full inversion for both aerosols and gases are carried out to compare the effect of the aerosol parameterization scheme on the retrieval accuracy of the atmospheric gases.

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Table 1. Some of the parameters employed to model the aerosols and the standard deviation in them.

Parameters	X	$m(z)$	$\sigma_X(z)$	r_C, km
Concentration of H_2O	C	ref. 36	100%	5
Concentration of SO_2	C	ref. 36	100%	5
The sulfuric acid aerosol	C	ref. 21	100%	3
	r_0	ref. 21	ref. 2	3
	σ	ref. 21	20%	3
Volcanic dust and meteoritic dust – 1	C	ref. 21	100%	3
	r_0	ref. 21	ref. 2	3
	σ	ref. 21	20%	3
Meteoritic dust – 2	C	ref. 21	200%	3
	r_0	ref. 21	100%	3
	σ	ref. 21	30%	3
All aerosol types	b	-3 (66% cases)	0.1	3
		-2 (34% cases)	0.8	3

C is the gas/aerosol concentration.
r_0 and σ are the parameters of the generalized lognormal size distribution (8).
$m(z)$ and $\sigma_X(z)$ are the mean and r.m.s. deviations of the parameters, X. r_c is the correlation radius.

Table 2. The eigenvalues of the covariance matrices, K_β and K_β^L

i, number	eigenvalue of K_β, km^{-2}	d_i of K_β, %	eigenvalue of K_β^L, km^{-2}	d_i of K_β^L, %
1	0.370-4	98.244776	574.05	98.12518
2	0.630-6	99.919194	10.02	99.83868
3	0.274-7	99.991955	0.855	99.98484
4	0.263-8	99.998941	0.823-1	99.99890
5	0.250-9	99.999604	0.533-2	99.99981
6	0.574-10	99.999757	0.760-3	99.99994
7	0.193-10	99.999808	0.146-3	99.99997

Table 3. Dependence of the absolute and relative r.m.s errors of the aerosol extinction approximation on the number of approximation parameters.

a) Linear parameterization (absolute error, km^{-1})			**b) Linear parameterization (relative error, %)**			**c) Logarithmic parameterization (relative error, %)**		
Number of parameters	Main data set	Independent data set	number of parameters	Main data set	Independent data set	number of parameters	Main data set	Independent data set
0	0.686E-3	0.960E-2	0	691.2	130.3	0	697.9	131.0
1	0.909E-4	0.149E-2	1	115.5	24.2	1	97.4	22.7
2	0.195E-4	0.574E-3	2	34.0	11.4	2	35.3	12.0
3	0.615E-5	0.350E-3	3	10.2	6.3	3	24.5	7.1
4	0.223E-5	0.172E-3	4	2.3	2.6	4	3.2	2.1
5	0.136E-5	0.714E-4	5	1.3	1.0	5	2.0	1.1
6	0.107E-5	0.435E-4	6	0.9	0.7	6	1.6	0.8
7	0.950E-6	0.408E-4	7	0.8	0.6	7	1.0	0.5

Table 4. Statistical characteristics of the errors of parameterizations.

Limit value [%]	Number of cases in which the error exceeds the limit value	
	Optimal (4 vectors)	quadratic
1	4807 (19.6%)	19519 (79.7%)
2	859 (3.5%)	14480 (59.1%)
3	172 (0.7%)	10666 (43.5%)
4	41 (0.2%)	7765 (31.7%)
5	9 (0.04%)	5559 (22.7%)
6	0	3943 (16.1%)
7	0	2655 (10.8%)
8	0	1769 (7.2%)

The total number of cases was 24,500.

Figure Captions

Figure 1. The first seven eigenvectors derived from the logarithm of the extinction spectra comprising the main data set.

Figure 2a. The spectral dependence of the relative error in the parameterization for different numbers of eigenvectors used in the approximation for one example simulation (#24 at an altitude is 48 km).

Figure 2b. The spectral dependence of the relative error in the parameterization for different numbers of eigenvectors used in the approximation for one example simulation (#2 at an altitude of 17 km).

Figure 3. The spectral dependence of the r.m.s. relative error for the main data set due to the logarithmic parameterization for different numbers of eigenvectors used in the approximation.

Figure 4. The spectral dependence of the aerosol extinction for the main and independent data sets. The vertical bars show the r.m.s. deviation for each data set.

Figure 5. The spectral dependence of the aerosol extinction for the three worst case approximations for each of the parameterizations. The curves are labeled with the simulation number, altitude and r.m.s. error.

Figure 6. An example of a spectrum and its approximation by the optimal eigenanalysis method for a case where the approximation error is large. Note the abrupt change in slope of the spectrum at 0.6 μm.

Figure 7. An example of a spectrum and its approximation by the quadratic method for a case where the approximation error is large. Note the changes in slope of the spectrum between 0.4 and 0.6 μm.

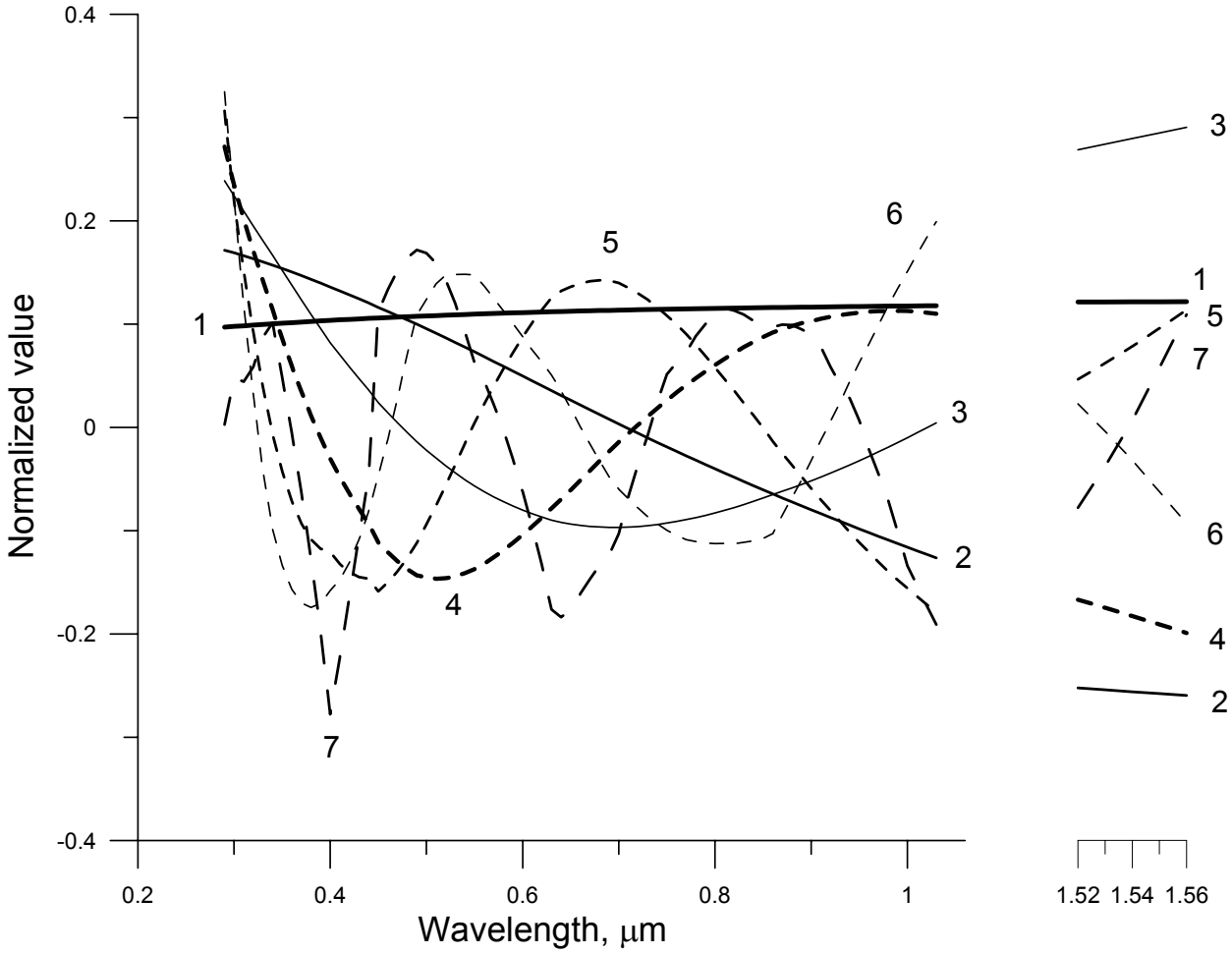


Figure 1. The first seven eigenvectors derived from the logarithm of the extinction spectra comprising the main data set.

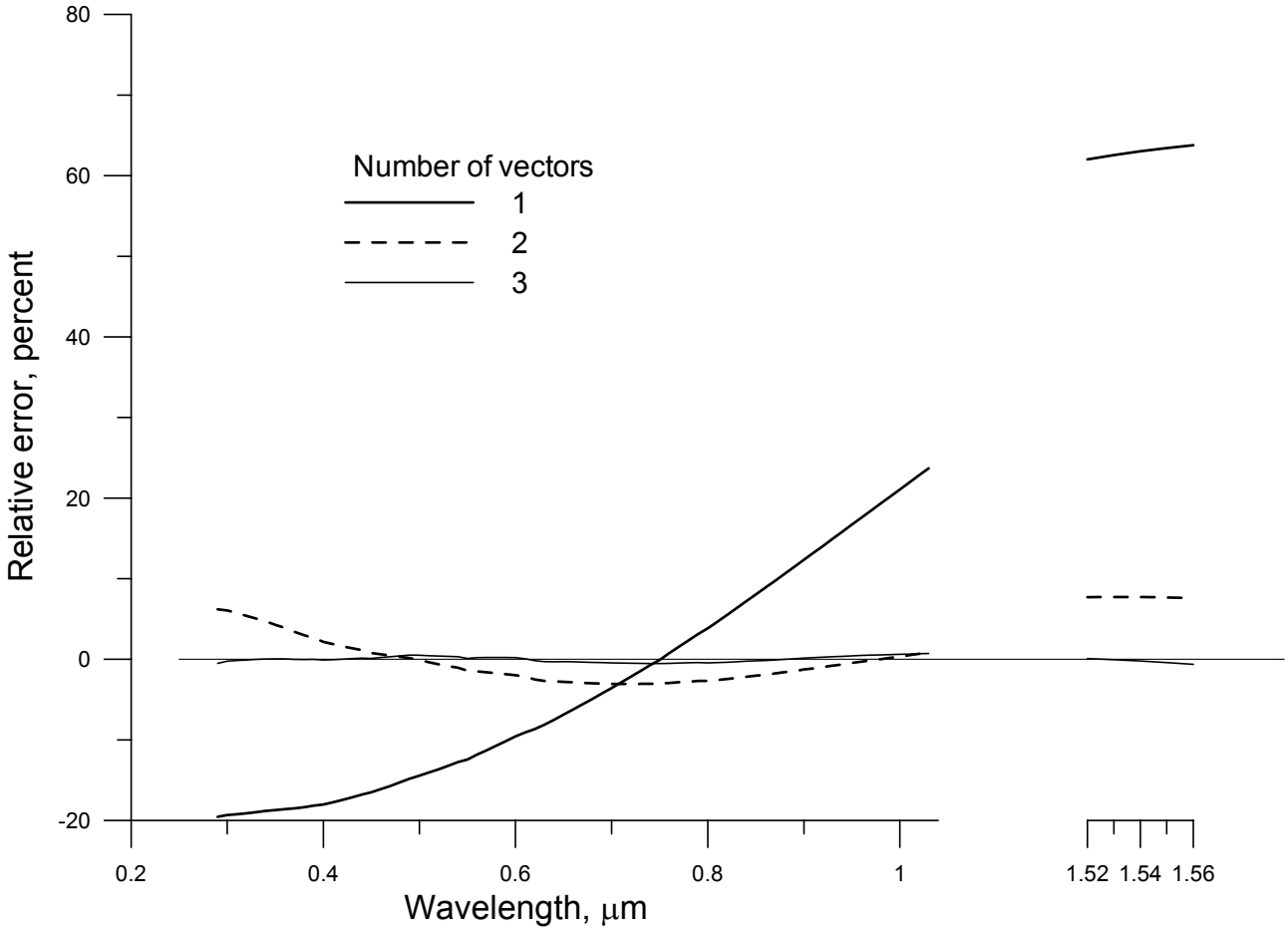


Figure 2a. The spectral dependence of the relative error in the parameterization for different numbers of eigenvectors used in the approximation for one example simulation (#24 at an altitude is 48 km).

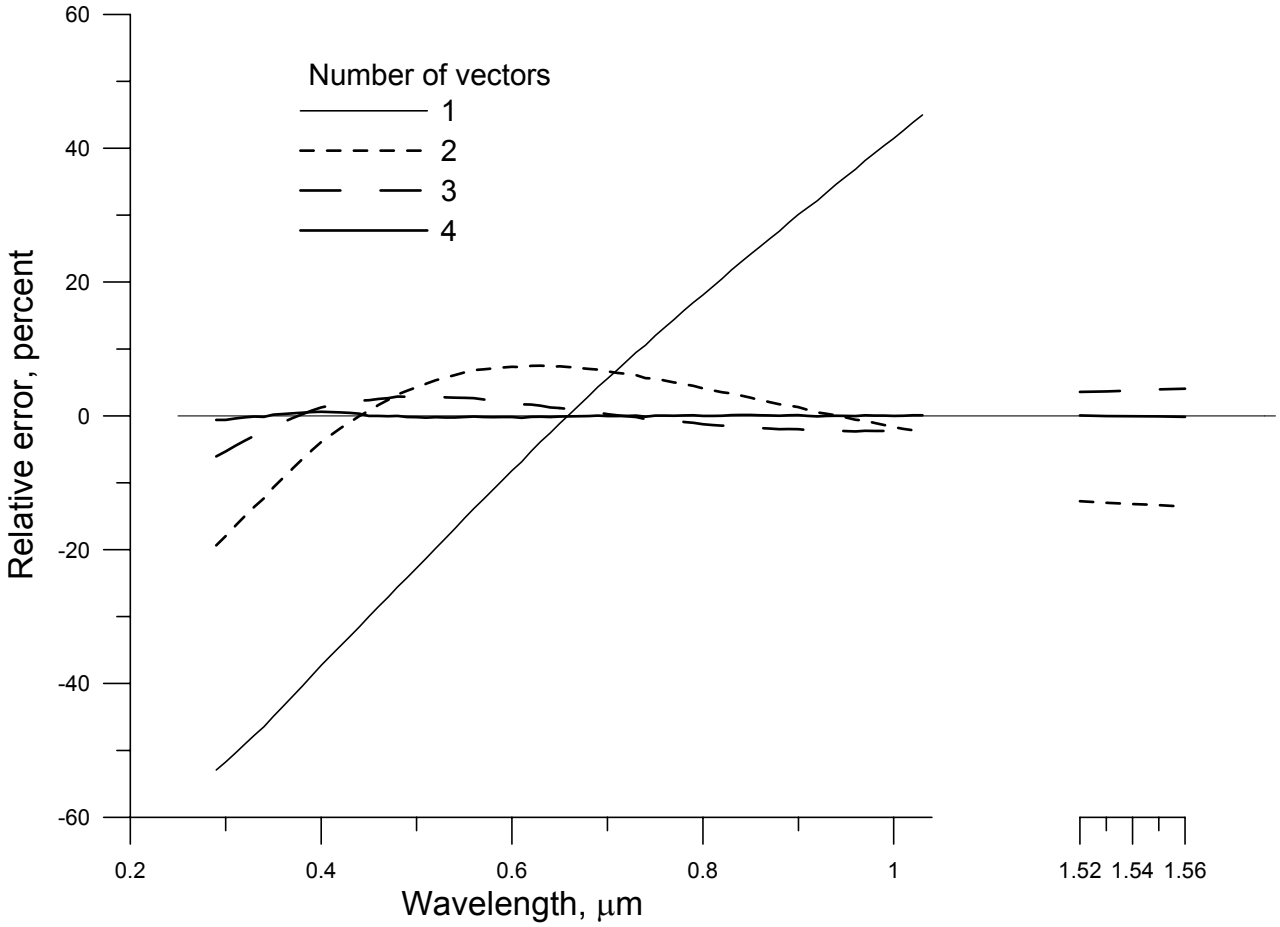


Figure 2b. The spectral dependence of the relative error in the parameterization for different numbers of eigenvectors used in the approximation for one example simulation (#2 at an altitude of 17 km).

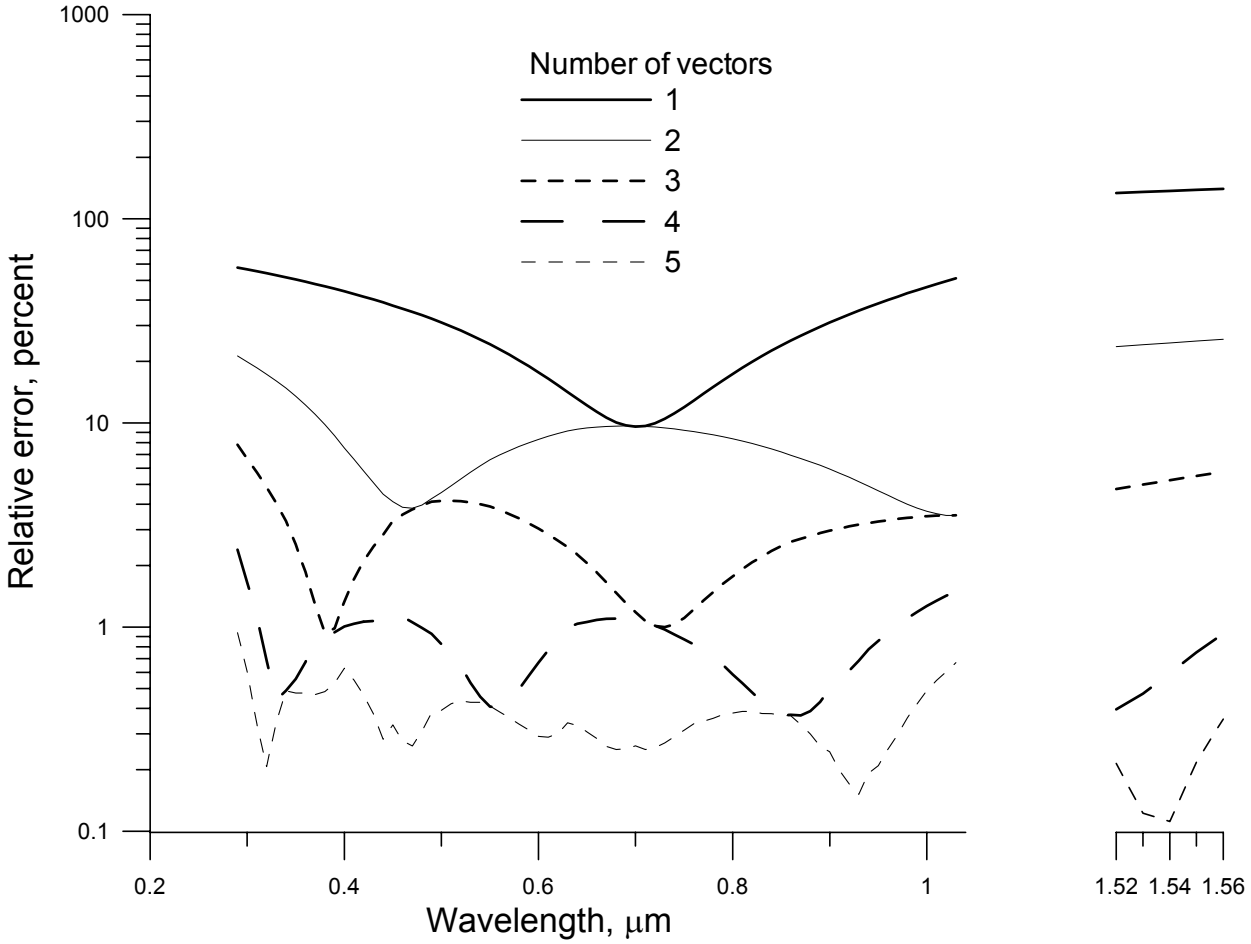
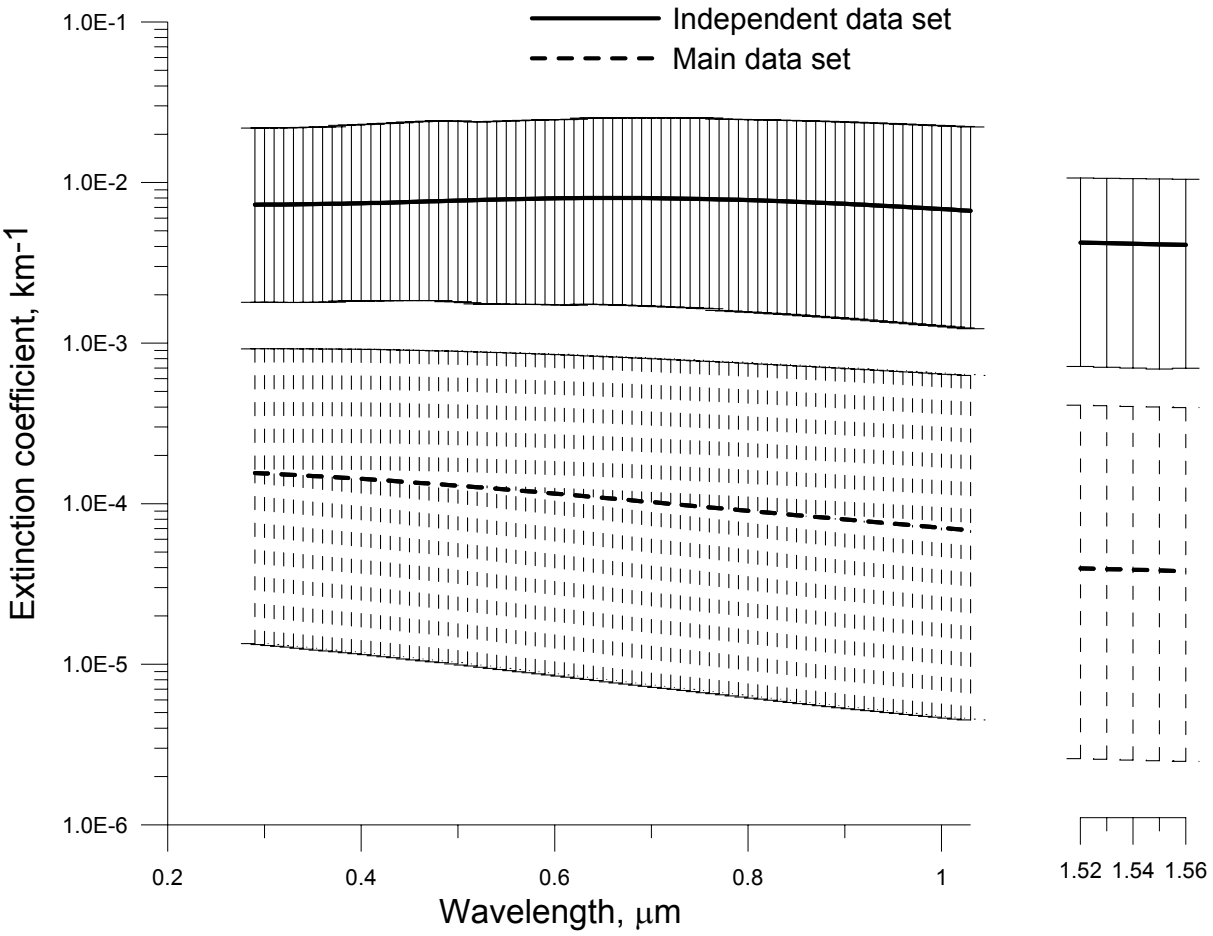


Figure 3. The spectral dependence of the r.m.s. relative error for the main data set due to the logarithmic parameterization for different numbers of eigenvectors used in the approximation.



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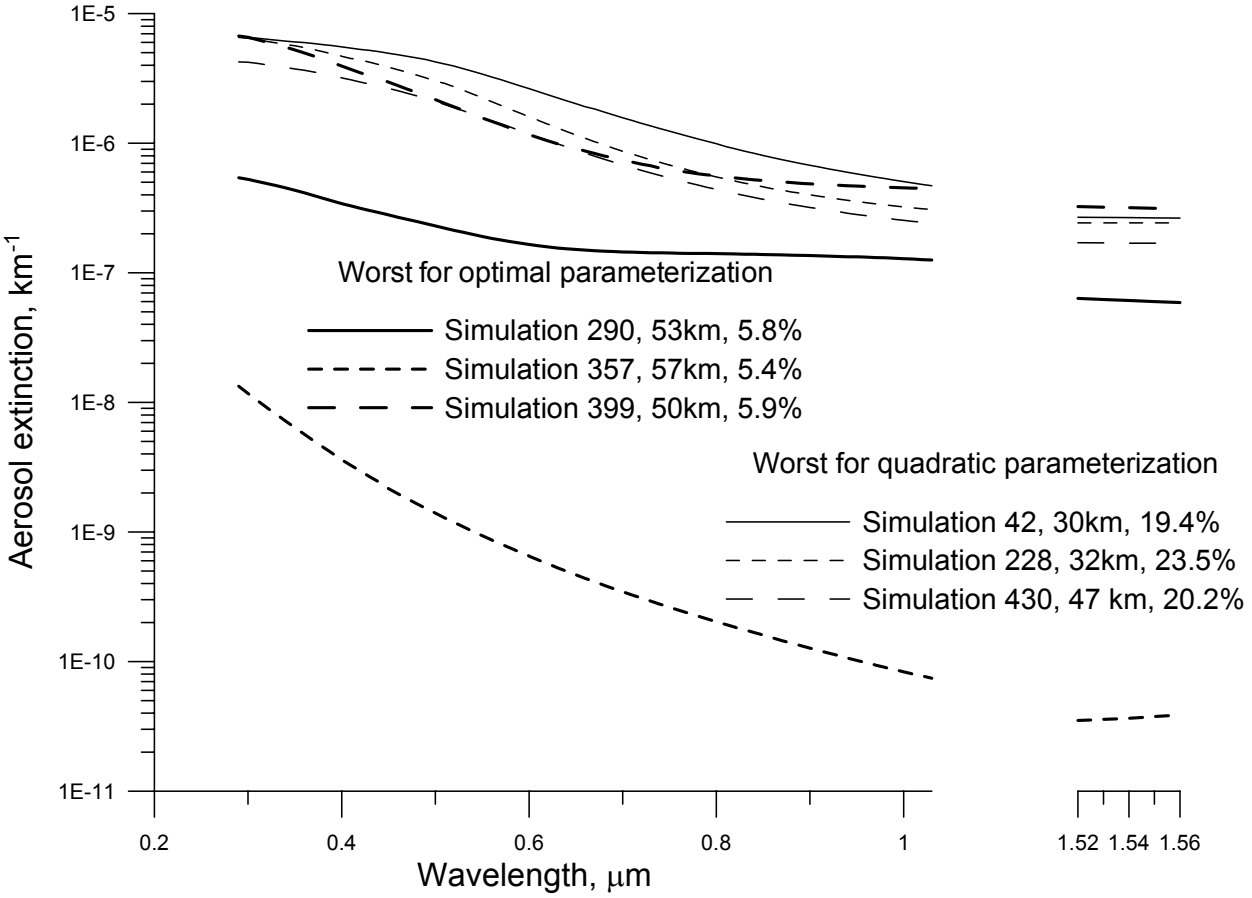

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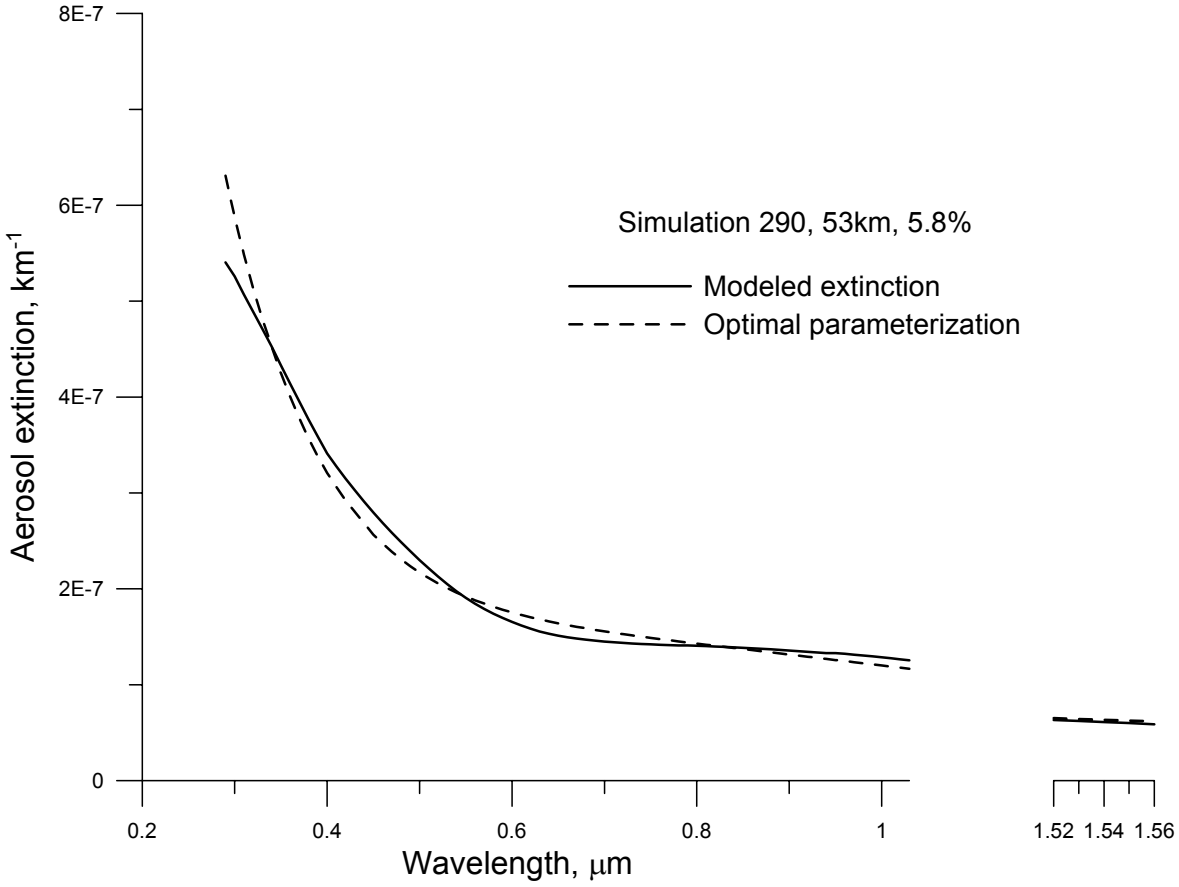

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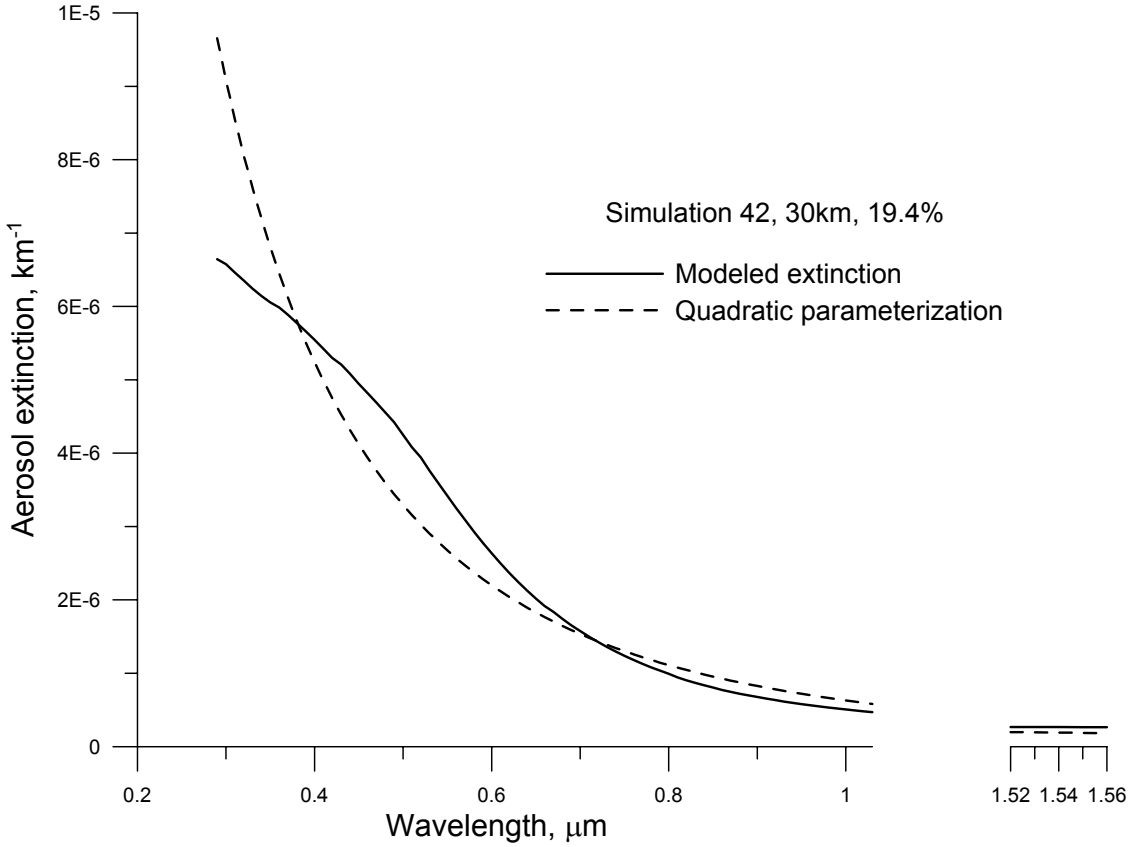

Figure 7. An example of a spectrum and its approximation by the quadratic method for a case where the approximation error is large. Note the changes in slope of the spectrum between 0.4 and 0.6 μm.