Retrieval of stratospheric aerosol size distribution from atmospheric extinction of solar radiation at two wavelengths

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The possibility of retrieving size distribution of aerosol particles in the stratosphere by measuring the extinction of solar radiation traversing the aerosol medium at two different wavelengths is explored in this work. This paper presents a parametric study of the effects of size distribution and composition of stratospheric aerosols on the value $R$, the ratio of extinction of solar radiation at wavelength 0.45 $\mu$m to the extinction at wavelength 1.0 $\mu$m. The aerosol size distributions under study are nine analytical expressions including most stratospheric aerosol models used by investigators. The aerosol compositions under study are the supercooled sulfuric-acid droplets, with different weight percentages of $H_2SO_4$ in the aerosol. It is found that $R$ is not very sensitive to either the composition or the radii limits of stratospheric aerosols under consideration but is quite sensitive to the value of the variable parameter governing the mode radius of the size distribution of aerosol particles. Based on the results of this parametric study, a method of retrieving the size distribution of aerosol particles in the stratosphere from the experimental results of the extinction of solar radiation at two wavelengths is proposed.

I. Introduction

Aerosol particles in the lower stratosphere received greater attention in recent years because research results have shown that they may play an important role in our complex environment. These particles may directly influence the heat budget of the atmosphere by absorbing, scattering, and emitting radiations depending on their optical properties. They may also indirectly influence the concentration of minor constituents in the atmosphere by changing the intensity of radiations available for production or destruction of ozone and other minor chemical species. The introduction of large amounts of gases and aerosols into the stratosphere by volcanic eruption and the diffusion of chemical species into the stratosphere due to mankind’s increasing activities will certainly affect the properties of these aerosols.

In past years both in situ and remote-sensing methods were used to obtain information about the optical and chemical properties of aerosol particles and their variation in space and time. In situ methods include balloon-borne particle counters, quartz-crystal microbalance impactor, filter impactor, wire impactor, and laser nephelometer. Remote-sensing methods include solar aureole almucantar, lidar measurements, and satellite. All these techniques (except for the airborne systems and satellite) give only a localized picture of the stratospheric aerosols for a relatively short interval of time. To monitor the stratospheric aerosols in a global and near-continuous base, two satellite experiments were started, and one of them is still in continuous operation. The first one is the Stratospheric Aerosol Measurement II (SAM II), whose instruments are onboard the Nimbus-7 satellite. The second one is the Stratospheric Aerosol and Gas Experiment (SAGE), whose instruments are onboard the Applications Explorer Mission-2 satellite. SAM II instruments have continued collecting data since the launch of the Nimbus-7 satellite on 23 Oct. 1978. However, SAGE instruments are not now in operation due to the failure of the electrical system but successfully collected data from 18 Feb. 1979 to 18 Nov. 1981.

The SAM II instruments are essentially sun photometers designed to measure the extinction of solar radiation at 1.0-µm wavelength during each sunrise and sunset event encountered by the satellite as it orbits the earth. Since the extinction of solar radiation by aerosol particles is a function of several parameters including...
Table 1. Size Distributions of Stratospheric Aerosols Used in This Study

<table>
<thead>
<tr>
<th>Model no.</th>
<th>Type</th>
<th>Formula</th>
<th>Value of fixed parameter</th>
<th>Variable parameter</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Lognormal (nonvolcanic)</td>
<td>$A \left( -\frac{\ln^2(r/r_g)}{2 \ln^2\sigma} \right) \sigma = 1.86 \quad r_g$</td>
<td>1</td>
<td>2</td>
</tr>
<tr>
<td>2</td>
<td>Exponential (nonvolcanic)</td>
<td>$A \exp\left( -\frac{r}{r_o} \right) \sigma = 2.0 \quad r_o$</td>
<td>3</td>
<td>4</td>
</tr>
<tr>
<td>3</td>
<td>Truncated power law</td>
<td>$\begin{cases} \sigma &lt; 0.1 , \mu m \ 0.1 , \mu m &lt; r &lt; 0.5 , \mu m \ \sigma &gt; 0.5 , \mu m \end{cases} \quad p \quad \gamma = 1 \quad b$</td>
<td>5</td>
<td>6</td>
</tr>
<tr>
<td>4</td>
<td>ZOLD-1 (nonvolcanic)</td>
<td>$A \exp\left( -\frac{\ln^2(r/r_m)}{2 \ln^2\sigma} \right) \sigma = 1.8 \quad r_m$</td>
<td>7</td>
<td>8</td>
</tr>
<tr>
<td>5</td>
<td>ZOLD-2 (postvolcanic insoluble)</td>
<td>$A \exp\left( -\frac{\ln^2(r/r_m)}{2 \ln^2\sigma} \right) \sigma = 2.0 \quad r_m$</td>
<td>9</td>
<td>10</td>
</tr>
<tr>
<td>6</td>
<td>ZOLD-3 (southern hemisphere)</td>
<td>$A \exp\left( -\frac{\ln^2(r/r_m)}{2 \ln^2\sigma} \right) \sigma = 1.72 \quad r_m$</td>
<td></td>
<td></td>
</tr>
<tr>
<td>7</td>
<td>Modified gamma-1 (haze H)</td>
<td>$A r^a \exp(-b r^a) \quad \alpha = 2 \quad \beta = 1 \quad \gamma = 1 \quad b$</td>
<td></td>
<td></td>
</tr>
<tr>
<td>8</td>
<td>Modified gamma-2 (AFGL background)</td>
<td>$A r^a \exp(-b r^a) \quad \alpha = 1 \quad \beta = 1 \quad \gamma = 1 \quad b$</td>
<td></td>
<td></td>
</tr>
<tr>
<td>9</td>
<td>Modified gamma-3 (AFGL volcanic)</td>
<td>$A r^a \exp(-b r^a) \quad \alpha = 1 \quad \beta = 0.5$</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

The purpose of this paper is to explore the possibility of retrieving aerosol size distribution from the extinction of solar radiation by these particles at a single wavelength. However, the SAGE instrument has four radiometric channels centered at 1.0-, 0.6-, 0.45-, and 0.385-\( \mu m \). The extinction of solar radiation by aerosols at two wavelengths, 1.0 and 0.45 \( \mu m \), can be obtained by the inversion method discussed in detail by Chu and McCormick.\(^1\) The purpose of this paper is to explore the possibility of retrieving aerosol size distribution from the extinction of solar radiation traversing the aerosol medium at those two wavelengths. Results of parametric study of the effects of aerosol size distribution and composition on the extinction of solar radiation at 0.45- and 1.0-\( \mu m \) wavelengths are presented and discussed. Based on the results of this parametric study, a method of retrieving the size distribution of aerosol particles in the stratosphere is proposed. Furthermore, the influence of the excess of large particles in the 1-\( \mu m \) radius region measured by Hofmann and Rosen\(^13\) on the proposed method is discussed.

### II. Model Assumptions

It has been shown that aerosol size distribution data can be represented by suitable analytic models.\(^14\) Recently, Russell et al.\(^15\) used nine different analytical types of size distribution for stratospheric aerosols in their optical model. Since these analytical expressions include most of the aerosol models used by different investigators and are based on experimental results, we adopted these models in this study. A summary of these models including the recommended values of the fixed parameters is listed in Table I. So that the constant \( A \) equals particles/cc in the formula for lognormal size distribution listed in Table I, this formula is slightly different from that used by Russell et al.\(^15\) Among these aerosol size distribution analytic models, the lognormal expression is probably the most popular one used for background stratospheric aerosol studies. It should be noted that, in each analytical expression, there are two unknowns, namely, the constant \( A \), which determines the number concentration of aerosol particles, and the variable parameter, which determines the mode radius of the aerosol size distribution. To eliminate one unknown we calculate \( R \), the ratio of the volume extinction coefficient of solar radiation by aerosol particles at wavelength \( \lambda = 0.45 \, \mu m \) to the volume extinction coefficient at wavelength \( \lambda = 1.0 \, \mu m \). The value of \( R \) is given by the expression

\[
R = \frac{\sigma_{0.45}}{\sigma_{1.0}} \frac{\int_{r_1}^{r_2} n(r)Q(\lambda = 0.45, x, m)\pi r^2 dr}{\int_{r_1}^{r_2} n(r)Q(\lambda = 1.0, x, m)\pi r^2 dr},
\]

where \( \sigma_{0.45} \) and \( \sigma_{1.0} \) are volume extinction coefficients of solar radiation at wavelengths 0.45 and 1.0 \( \mu m \), respectively; \( n(r) \) is the aerosol size distribution; \( Q(\lambda, x, m) \) is the Mie extinction efficiency which is a function of...
Fig. 1. Ratio of aerosol extinction coefficient at $\lambda = 0.45 \mu m$ to aerosol extinction coefficient at $\lambda = 1.0 \mu m$ as a function of the variable parameter in the aerosol size distribution $n(r)$ for different aerosol compositions. Solid curves are results obtained with $r_1 = 0.1 \mu m$, $r_2 = 1.0 \mu m$ in Eq. (1). Dotted curves are results obtained with $r_1 = 0.01 \mu m$, $r_2 = 1.0 \mu m$. Dashed curves are results obtained with $r_1 = 0.1 \mu m$, $r_2 = 3.0 \mu m$. $\bigcirc$ are results obtained from aerosols of 50% $H_2SO_4$, $\square$ are results obtained from aerosols of 75% $H_2SO_4$, and $\Diamond$ are results obtained from aerosols with 84.5% $H_2SO_4$. Curves with $\bigtriangleup$ show extinction coefficients calculated with constant $A = 1$ as a function of the variable parameter. Figures (a)-(l) correspond to results obtained from analytical models 1-9 listed in Table I, respectively.
Fig. 1. See caption on page 1641.
wavelength $\lambda$, size parameter $x = (2\pi r)/\lambda$, where $r$ is the radius of the particle assumed to be spherical, and refractive index $m$, which in turn is also a function of wavelength $\lambda$ and composition of the aerosol particle. It is obvious that $R$ is independent of the value $A$ but in general depends on the variable parameter in the aerosol size distribution expression $n(r)$, the values of $r_1$ and $r_2$, and the composition of the aerosol particles which determines the value of $m$.

It is generally accepted that the stratospheric aerosol particles are primarily composed of sulfuric acid and water, with $\sim 75\%$ by weight of $\text{H}_2\text{SO}_4$ in the aerosol. A recent paper by Turco et al. has reviewed the controversy of whether ammonium sulfate is present in stratospheric aerosols. They concluded that only small quantities of ammonium ions may be present. In this parametric study we consider three different compositions of stratospheric aerosols, namely, pure sulfuric-acid aerosols with weight percentages of $\text{H}_2\text{SO}_4$ in the aerosols being 50, 75, and 84.5%. The weight percentages of 50, 75, and 84.5% are chosen because their refractive indices for different wavelengths at 300 K are given by Palmer and Williams. The refractive indices at a stratospheric temperature of 220 K are obtained by applying the Lorentz-Lorenz formula. The values of refractive indices used in this study are listed in Table II. Values shown in Table II are for the real part of the refractive index only; since the imaginary parts of all the refractive indices are $\sim 10^{-6}$ or less, we assume the imaginary part of the refractive indices is zero in our calculations.

III. Discussions of the Calculated Results

The values of $R$ as a function of the variable parameter for four types of aerosol composition and different values of $r_1$ and $r_2$ are shown in Figs. 1(a)–(i). Different figures correspond to different expressions of $n(r)$. In each figure solid curves are obtained by using $r_1 = 0.1 \mu m$, $r_2 = 1.0 \mu m$; dotted curves are obtained by using $r_1 = 0.01 \mu m$, $r_2 = 1.0 \mu m$; dashed curves are obtained by using $r_1 = 0.1 \mu m$, $r_2 = 3.0 \mu m$. For the sake of clarity only those portions of the dotted and dashed curves that deviate from the solid curve obtained from aerosols of 50% $\text{H}_2\text{SO}_4$ are shown. The deviations of the dotted and dashed curves from the solid curves corresponding to other aerosol compositions are similar to those shown in these figures.

From the plotted curves shown in these figures it can be seen that $R$, the ratio of aerosol extinction coefficients at two wavelengths, is a monotonic function of the variable parameter. Furthermore, $R$ is quite sensitive to the variable parameter when $R$ is much larger than 1. Consequently, the experimental value $R$ can be utilized to estimate the aerosol size parameter from the plotted curves.

Except in the case of truncated power law as shown in Fig. 1(c), the value $R$ is in general not very sensitive to the composition of aerosol particles, particularly when the weight percentages of $\text{H}_2\text{SO}_4$ in the aerosol is $\sim 75\%$. As a result, the value of the variable parameter in the expression of aerosol size distribution can be determined even if we do not know the exact composition of aerosol particles.

Changing the upper or lower limit of the aerosol size distribution will in general affect the value $R$. However, the effect on $R$ is small in comparison with the change in variable parameters. This fine feature of the value $R$ enables us to estimate the size distribution of aerosol particles in the stratosphere without exactly knowing its size range.

In addition to the values of extinction ratio $R$, the values of extinction coefficient at $\lambda = 1.0 \mu m$, $\sigma_{1,0}$ calculated with constants $A$ in the size distribution expression equal to unity, with $r_1 = 0.1 \mu m$ and $r_2 = 1.0 \mu m$, and assuming aerosols are 75% $\text{H}_2\text{SO}_4$ and 25% $\text{H}_2\text{O}$ mixtures, are also presented in Figs. 1(a)–(i). After the values of the variable parameter have been determined from the measured extinction ratio, the expected extinction coefficient with constant $A = 1$ can be determined from the plotted curve in the same figure. The value of the constant $A$ equals the ratio of the measured value $\sigma_{1,0}$ to the expected extinction coefficient obtained in the previous step. A complete knowledge of the aerosol size distribution including the constant $A$, which
governs the aerosol number concentration, and the variable parameter, which governs the mode radius, is then obtained.

To illustrate the proposed technique let us consider an example with measured results $\sigma_{1.0} = 1.1 \times 10^{-4}$ km$^{-1}$ and $\sigma_{0.45} = 4.4 \times 10^{-4}$ km$^{-1}$. In this case, $R = 4.0$. If we assume a background stratospheric aerosol with a lognormal size distribution, the mode radius $r_m$ can easily be determined to be $\sim 0.062 \mu$m from the solid curve in Fig. 1(a). The expected extinction coefficient with $A = 1$ is then determined from the curve with triangle symbols in the same figure. This value turns out to be $\sim 1.1 \times 10^{-6}$ km$^{-1}$. The ratio between the measured value $\sigma_{1.0}$ and this expected value is 10. Since the constant $A$ for the lognormal size distribution equals the aerosol number concentration, we conclude that the stratospheric aerosols producing the measured results have a number concentration of 10 particles/cm$^3$.

Since the extinction profiles at $\lambda = 1.0$ and 0.45 $\mu$m can be retrieved to $\sim 10\%$ accuracy between 13 and 24 km extended over the full altitude range of the stratospheric aerosol layer, let us assume that the uncertainty in $R$ is 10%. The mode radii for $R = 4.4$ and 3.6 are 0.055 and 0.07 $\mu$m, respectively. Consequently, the expected error of the mode radius determined from the SAGE result is $\sim 13\%$.

On the other hand, if we assume the measurement was obtained after volcanic eruption, we may decide to choose the ZOLD-2 aerosol size distribution listed as model 5 in Table I. The variable parameter $r_m$ is then determined to be 0.055 $\mu$m in Fig. 1(e). The expected extinction coefficient with $A = 1$ is determined to be $1.6 \times 10^{-6}$ km$^{-1}$ from the same figure. The value of constant $A$ in the ZOLD-2 expression is then calculated to be $\sim 70$.

IV. Concluding Remarks

The variations of the value $R$, the ratio of extinctions of solar radiation at wavelengths $\lambda = 0.45 \mu$m and $\lambda = 1.0 \mu$m with the variable parameter for different compositions, and the lower and upper limits of the aerosol radii under consideration for nine commonly used types of aerosol size distribution are presented graphically in Figs. 1(a)–(i). In addition, the expected extinction coefficient with constant $A = 1$ in the aerosol size distribution as a function of the variable parameter is also presented in these figures. From the plotted curves it can be seen that, although the uncertainty of aerosol composition and the values of $r_1$ and $r_2$ may affect the value $R$, in general it is not sensitive to these parameters, especially for large values of $R$ but is very sensitive to the value of the variable parameter. Consequently, the experimental value $R$ can be used to estimate the value of the varied parameter in the assumed analytical expression even without prior knowledge of the composition and size range of the aerosol particles responsible for the extinction of solar radiations at these two wavelengths. Once the value of the variable parameter is determined, the expected extinction coefficient with constant $A = 1$ can be determined from the same figure. The value $A$ governing the aerosol number concentration can then be easily calculated by taking the ratio of the measured $\sigma_{1.0}$ to the expected extinction coefficient determined previously.

The plotted curves presented in Figs. 1(a)–(i) can be utilized to quickly estimate the aerosol size distribution from experimental results of $\sigma_{1.0}$ and $\sigma_{0.45}$. The uncertainty produced by aerosol composition and the values of $r_1$ and $r_2$ can also be estimated from the presented curves.

Recently, Hofmann and Rosen used a specially designed large particle counter to measure concentrations of particles with radii between 1 and 2 $\mu$m. Their results indicated that, although the concentration of smaller particles can be fitted with a lognormal curve, the concentrations of these particles with radii >0.95 $\mu$m are much larger than that suggested by the lognormal fitting of smaller particles. To study the effect of these large particles on the expected values of $R$ we have calculated the values of $R$ with and without the presence of larger particles as reported by Hofmann and Rosen on their 15 Jan. 1981 measurements. The calculated results are listed in Table III. The inferred mode radii from the values of $R$ are also shown in this table. From this table it can be seen that the presence of large particles in the stratosphere will decrease the extinction ratio by a value <15%, and the estimated mode radius will be increased by a value less than $\sim 17\%$. A recent report by Deepak et al. has shown that the presence of these larger particles can increase the backscatter at CO$_2$ wavelength ($\lambda = 10.6 \mu$m) by more than 1 order of magnitude. In comparison, the presence of larger particles in the stratosphere has a much more significant effect on the backscatter at CO$_2$ wavelength than on the aerosol extinctions and inferred mode radii from satellite measurements at visible and near-infrared wavelengths.

It should be noted that the proposed technique is

| Table III. Comparison of the Expected Extinction Ratios $R$ Calculated With and Without the Presence of Large Particles and Their Corresponding Mode Radii $r_m$ Determined By the Proposed Technique |
|---|---|---|---|---|
| Height (km) | Without large particle | With large particle | Without large particle | With large particle | Percentage difference |
| 13 | 3.97 | 4.00 | 0.062 | 0.050 | For $R$ For $r_m$ |
| 16 | 4.33 | 4.33 | 0.068 | 0.058 | 13.63 | 17.24 |
| 19 | 5.00 | 4.27 | 0.072 | 0.068 | 16.00 | 16.0 |

* The aerosol size distributions used in these calculations are taken from Hofmann and Rosen on their 15 Jan. 1981 measurements.
basically applicable to aerosol size distributions with a single mode. It is possible to extend this technique to aerosols with a bimodal character if we know or can assume the properties of aerosols in the second mode of larger particles. However, even if we do not know the properties of these larger particles, the proposed technique can still be used but with a possible error of <20% as discussed previously.

It is possible to deduce the properties of larger aerosols by measuring the aerosol extinction at one or two additional wavelengths. Such possibilities, as well as the possibility of using other measurement techniques including choosing wavelengths other than those used by SAGE in order to get optimum results, are currently under investigation and will be reported later. In the meantime we have demonstrated that this retrieval technique can be applied to obtain useful aerosol size distributions in the stratosphere from the available satellite experimental data.

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