Calculation of Aerosol Optical Scattering Characteristics

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Abstract The solution of aerosol scattering characteristics has always been a hot topic in the field of atmospheric science owing to its basic status. The difficulty of solving aerosol scattering characteristics is increased. Therefore, there is not yet a unified scattering theory which can be used to calculate various aerosol particles. In this paper, firstly, the basic principles and research progress of existing methods for solving the scattering characteristics of aerosol light are summarized. Secondly, the advantages and disadvantages of these solution methods are compared. Finally, the problems existing in the current research and the future development prospects are analyzed.

1. Introduction

The atmosphere is the environment on which humans depend with complicated composition. In addition to the most basic gas components, various solid, liquid and mixed particles are also widely found in every corner of the Earth's atmosphere. These multiphase systems consisting of particles suspended in the atmosphere ranging from 0.001\(\mu\text{m}\) to 100\(\mu\text{m}\) are called aerosols\textsuperscript{[1]}. There are two main sources: one is nature's own products, such as clouds, fog, sand, sea salt, volcanic ash, microorganisms, etc. in the atmosphere; second, human production and life, such as engine tail smoke, artificial dust, industrial and domestic emissions, etc.

As an important part of the atmosphere, aerosols are important factors affecting the climate and the environment. Atmospheric aerosol particles can directly or indirectly affect the climate by scattering and absorbing solar radiation, as a cloud of condensation nuclei\textsuperscript{[2]}, and even directly lead to air pollution, affecting human living environment. Optical parameters such as absorption and scattering factors of aerosol particles are important factors for evaluating atmospheric pollution and studying the climatic effects of aerosol radiation. Therefore, the study of aerosol light scattering characteristics has been the focus of international research and cutting-edge topics \textsuperscript{[3]}. From the literature analysis, the methods for studying this problem mainly include two categories: actual observation and simulation.

Observing research can get the most realistic data, and has the most accurate guiding significance for obtaining correct results. At present, many units have established a database describing aerosol light scattering characteristics based on observation data of equipment such as laser radar. However, actual observations are limited by the nature of the site location and its computational statistical properties, and do not give the scattering characteristics of all aerosols. The simulation calculation does not require large observation equipment, and can build a rich and
diverse model, which has gradually become an important method to study the light scattering characteristics of aerosol particles.

2. Calculation of aerosol optical scattering characteristic

The simulation of aerosol optical properties has been continuously developed. The research object has evolved from the initial homogeneous spherical particles to ellipsoidal, cylindrical, eccentric spherical, layered spherical clusters and arbitrary complex shapes. The research methods have also gone from simple to complex. According to the applicable conditions, they can be roughly divided into three categories, analytical method, approximation method and numerical method. Analytical method mainly refers to Mie scattering theory; approximation method includes Rayleigh approximation and geometric optical approximation; numerical method can be divided into three types based on basis function expansion, volume integral equation and scattering method based on micro element method. The characteristics of various methods are compared as shown in Table 1.

<table>
<thead>
<tr>
<th>Method</th>
<th>Advantage</th>
<th>Disadvantage</th>
</tr>
</thead>
<tbody>
<tr>
<td>Analytic method</td>
<td>High computational accuracy</td>
<td>The surface of particles must conform to the surface of the corresponding coordinate system, Small scope of application</td>
</tr>
<tr>
<td>Approximate method</td>
<td>Simple calculation, Easy to implement</td>
<td>Rough theoretical model, Low calculation precision, Limited scope of application</td>
</tr>
<tr>
<td>Numerical method</td>
<td>Wide range of application, Higher computational accuracy</td>
<td>Computational complexity, Low efficiency, Involving repeated computation</td>
</tr>
</tbody>
</table>

2.1 Mie scattering calculation model

The Mie scattering calculation model is a homogeneous spherical particle scattering model independently proposed by the German physicist Mie and the Danish physicist Lorenz. The model obtains the analytical solution of the light scattering properties of spherical particles incident on a plane wave by accurately solving Maxwell's electromagnetic equations. Based on this theoretical model, the beam scattering law of any kind of spherical uniform particles can be studied. The Mie scattering calculation model is currently the most widely used particle scattering calculation model, especially for dealing with the scattering of wavelength-level particles with unparalleled precision.

However, the Mie scattering model is only suitable for spherical smooth particles with a smooth surface, and the scope of application is very limited. Hulst[4] discussed the Mie scattering theory in detail in the book Light scattering by small particles. Later, some scholars have improved it and further established improved theories for different conditions, the inherent limitations of the Mie scattering model are still difficult to overcome. In order to study non-spherical particles using the Mie scattering model, the non-spherical particles are usually equivalent to spherical particles. The radius of the equivalent spherical particles is generally obtained by the equal volume method, the equal surface area method and the equivalent volume to area ratio. However, a large number of experiments have proved that this approximation will cause a large deviation between the simulated value of the Mie theory and the scattering characteristics of the non-spherical aerosol. Some literatures[5] have shown that the error of calculating the scattering energy of non-spherical particles is as high as 60%. Some scholars[6] calculated the scattering properties of different types of aerosol
particles by equal volume and equal surface area approximation, and discussed the effect of particle shape distortion and properties on the equivalent Mie scattering error.

In summary, the Mie scattering theory is highly accurate in the processing of scattering of spherical particles at the wavelength level, but the size range of common aerosol particles is very large (0.001 μm~100μm), and the shape is also very different, so there are more scattering of particles. The characteristics are not suitable for studying with Mie theory. For particles with large differences in size from wavelength, the approximate scattering model can solve its scattering characteristics better.

2.2 Approximate calculation model for aerosol particle scattering characteristics

The ratio of the particle radius ($r$) to the radiation wavelength ($\lambda$) is the key to the degree of scattering. The dimensionless scale parameter $\alpha=2\pi r/\lambda$ is usually used as a reference standard for discriminating which research method is used. When $\alpha\ll1$, the incident light wavelength is much larger than the particle size. The commonly used model is Rayleigh scattering approximation and Rayleigh-Gans-Stevenson approximation model. When the particle radius and the radiation wavelength are about the same magnitude, the Mie scattering model is commonly used. When $\alpha\gg1$, the incident light wavelength is much smaller than the particle diameter, a geometric optical model and an abnormal scattering approximation model are usually used.

2.2.1 Rayleigh scattering approximation model

The Rayleigh Approximation Model (RA) is mainly for particles with a particle size much smaller than the wavelength of the incident light. At this time, the scattering intensity is inversely proportional to the fourth power of the incident light wavelength, that is, the shorter the wavelength, the stronger the scattering. For simple shape particles, the RA model has a completely analytical solution; for complex shape particles, the RA model solution can be obtained by simply solving the integral equation of the polarization vector. Bohren and Jones and others have improved the RA approximation model and expanded its scope of application.

The Rayleigh-Gans-Stevenson Approximation Model (RGA) is an enhanced version of the Rayleigh scattering approximation model. The main improvement is to increase the order of the basis function from 2 expansions of the scale parameter to 4 expansions. In this model, each micro-element in the scatter is only excited by the incident field and is scattered, independent of other factors. Compared to the RA approximation, the range of applicable scale parameters has increased. After many scholars continue to expand, the current applicable shape of the model includes ellipsoids, cylinders, aniso-tropic particles and particles with Gaussian random surfaces.

2.2.2 Abnormal diffraction approximation model

The Anomalous Diffraction Approximation (ADA) model was originally used to calculate the particle extinction cross section. After years of development, the model can be applied to both complex shaped particles and semi-empirical simulations of smaller scale parameters and larger refractive index particles. Light scattering characteristics. At present, the particle shapes applicable to the ADA model mainly include prisms, hexagonal ice crystals, ellipsoids, finite cylinders, and cubes. For different shapes of particles, the accuracy of ADA model simulation is not consistent. LIU[7] found that the simulation accuracy of ADA for randomly oriented ellipsoidal particles is higher than that of spherical particles. And the absorption coefficient simulation error of ADA increases as the imaginary part of the complex refractive index increases.
2.2.3 Geometric optical approximation model

Geometric Optics Approximation (GOA) is widely used for the simulation of light scattering properties of particles with scale parameters much greater than one. The basic idea is to simulate the refraction and reflection process of light inside and outside the particle through Snell's law and Fresnel equation. The diffraction process is processed by Fraunhofer diffraction principle, and the contribution of each light in a certain solid angle interval is averaged. Scattered light energy distribution\textsuperscript{[8]}. This method has the advantages of fast calculation speed and simple program structure. However, Fraunhofer diffraction does not explain the polarization properties of electromagnetic fields and leads to discontinuous distribution of scattering energy. Therefore, an improved geometric optical approximation method is developed to avoid solid angle averaging of reflected and refracted energy. But the traditional GOA method does not hold strong absorbing particles. By using the imaginary part of refractive index to characterize the absorption characteristics of particles, it can realize the geometric optical approximation.

In general, the approximate solution model of aerosol particle scattering characteristics has its specific scope of application. Some methods even require physical properties of aerosol particles and incident fields around the particles. Therefore, it is necessary to clarify the scope of application and application conditions of various methods. The applicable scope, advantages and disadvantages of the above several approximate models are summarized in Table 2.

Table 2. Approximate calculation model of scattering characteristics of non-spherical particles.

<table>
<thead>
<tr>
<th>Model</th>
<th>Inventor</th>
<th>Particle types</th>
<th>Advantage</th>
<th>Disadvantage</th>
</tr>
</thead>
<tbody>
<tr>
<td>RA</td>
<td>Rayleigh</td>
<td>The particle size is far less than the wavelength. Only some simple shapes have analytic solutions.</td>
<td>There is a complete analytic solution for simple shape. Only a simple numerical solution is needed for complex shape particles.</td>
<td>Particles are homogeneous, the scale is far less than wavelength. The incident field inside and around the particle is an approximate electrostatic field.</td>
</tr>
<tr>
<td>RGA</td>
<td>Stevenson</td>
<td>The particle size is far less than the wavelength. Optical soft particles.</td>
<td>It is an analytical solution. The scope of application is larger than that of Rayleigh.</td>
<td>It is only suitable for particles with a size far less than the wavelength.</td>
</tr>
<tr>
<td>ADA</td>
<td>Van de Hulst</td>
<td>Applicable to any shape of a particle in theory.</td>
<td>Suitable for large scale optical soft particles. Fast and accurate algorithm.</td>
<td>Only suitable for particles with the radius far greater than wavelength. Unable to calculate the pattern of scattering phase function.</td>
</tr>
<tr>
<td>GOA</td>
<td>Liou et al</td>
<td>Applicable to any shape of a particle in theory. Mainly used for ice crystal particles.</td>
<td>The concept is simple and the realization is convenient. The only practical method to solve the large particle scattering.</td>
<td>Not suitable for the simulation of light scattering of small particles. Must pay great attention to the lower limit of the use of the model.</td>
</tr>
</tbody>
</table>

2.3 Numerical Solution Model for Scattering Characteristics of Aerosol Particles

The numerical solution model of scattering characteristics refers to a method of obtaining the electromagnetic scattering characteristics of particles by directly solving electromagnetic wave propagation equations under certain boundary conditions, such as Maxwell equations and...
Helmholtz equations. The general problem with this type of method is that the calculation is relatively complicated, but with the rapid development of computer performance and the emergence of various fast algorithms in computational mathematics and computational physics, the ability to solve numerical methods has been greatly improved. Many numerical methods have been easily solved, and numerical methods have been greatly developed. The following is a brief introduction to several of the most widely used numerical methods.

2.3.1 T matrix method

The basic principle of T Matrix Method (TMM) is to use the vector ellipsoid wave function to expand the incident field, the scattering field and the internal field, and calculate it by expanding the boundary condition method. The T matrix is obtained by combining the transformation matrix of the incident field into the internal field and the transformation matrix of the internal field to the scattering field. Once T is obtained, the scattering cross section in any direction can be obtained, and the scattering characteristics of the aerosol particles can be solved. The accuracy of the T-matrix method has been extensively verified and is widely used. Wei Xiaodong\[9\] combined the T matrix method with the geometrical optics method to calculate the optical properties of the ellipsoid dust particles, and discussed the effect of the shape on its optical properties. Mishchenko\[10\] used the mathematical convergence method to overcome the instability of numerical calculation when the particle rotation axis is too long. At present, T matrix method is one of the best and most widely used methods for strictly calculating the light scattering effect of resonant non-spherical particles.

2.3.2 Method of Moments

Method of Moments (MoM) was first proposed by Harrigton and gradually evolved into an accurate numerical method commonly used in electromagnetic calculations. MoM is theoretically an accurate solution method that is widely used for scattering calculation of rough surfaces without special boundary conditions. However, its biggest drawback is that it has a large amount of unknowns and a large amount of calculation, so it is only used to deal with scattering problems in low frequency or resonant regions. Cui Zhiwei and Han Yiping\[11\] studied the Gaussian scattering problem of particles of arbitrary shape based on this method. In recent years, many scholars are devoted to the calculation efficiency of MoM by reducing the filling time of the impedance matrix, accelerating the product of the matrix and reducing the matrix equation dimension. The advantage of MoM is that it is suitable for particles with arbitrary shape, non-uniformity and anisotropy, and the physical concept is simple. However, MoM also has problems such as low numerical precision, complicated calculation. So it is usually used to calculate the scattering problem of small particles.

2.3.3 Finite Element Method

Finite Element Method (FEM) is a frequency domain differential equation method based on the variational principle and weighted residual method. It was first proposed by Morgan et al. The finite element method completely eliminates the effect of solid angle dispersion on the scattering characteristics. It has now evolved into a method that can be used for particle shape simulation of arbitrary shapes. The finite element method simulates a variety of complex shapes with the flexibility of discrete elements in the solution process. However, due to the need to artificially introduce an absorbing boundary condition, the efficiency is not high. Li Fangxia\[12\] used the finite element method to theoretically analyze the nonlinear aerosol dynamic equation. At present, the finite element method in the Cartesian coordinate system is usually used to deal with the isotropic scattering problem. The method of dealing with the linear anisotropic scattering problem is studied in [13].
2.3.4 Finite Difference Time Domain

The Finite Difference Time Domain (FDTD) was proposed by Yee in 1966. It is a scattering model based on the idea of micro-element. FDTD takes the time difference of the time domain Maxwell curl equation, discrete way of spatially alternating sampling of magnetic and electric fields, convert the curl equation into a set of difference equations, thereby obtaining a numerical solution. The method has the advantages of simple algorithm, avoiding the singular kernel problem often existing in the integral equation method, easy to implement programming and parallel operation, and capable of dealing with scattering problems of complex shapes and non-uniform small particles, and has high accuracy. The main disadvantage is that under the condition of improving the accuracy, the time step will be shorter, resulting in too much calculation, and the accumulated error will be accumulated for long time simulation and large target calculation. Through the improvement of Tamlolve\cite{14}, FDTD can be applied to the scattering simulation of non-uniform particles with arbitrary complex shapes. Multi-Resolution Time Domain (MRTD) and Pseudo Spectral Time Domain (PSTD) have been proposed, which improve the computational efficiency and calculation accuracy of FDTD under some conditions. In general, the FDTD method is more suitable for the light scattering calculation of small particles, its further promotion has been limited because of repeated calculations.

With the continuous advancement of computer technology and the continuous development of various methods in electromagnetics and mathematics, numerical solutions have gradually become the mainstream solution for the scattering characteristics of aerosol particles. There are many kinds of numerical solutions, each with its own merits. The performance comparison, computational complexity, advantages and disadvantages of several common methods are shown in Table 3.

<table>
<thead>
<tr>
<th>Model</th>
<th>Size</th>
<th>Particle types</th>
<th>Complexity</th>
<th>Advantage</th>
<th>Disadvantage</th>
</tr>
</thead>
<tbody>
<tr>
<td>TMM</td>
<td>$\alpha &lt; 180$</td>
<td>Rotationally symmetric particles. Multilayered scatterer. Sphere particle cluster.</td>
<td>$O(x^3) \sim O(x^4)$</td>
<td>High precision, fast speed, especially suitable for rotating symmetric particles. High credibility, widely tested.</td>
<td>It is easy to find the instability of numerical solution with large particle size, extreme shape or larger complex refractive index.</td>
</tr>
<tr>
<td>MoM</td>
<td>Small particles</td>
<td>Inhomogeneous particles of arbitrary shape</td>
<td>$O(x^6)$</td>
<td>Applied to particles of arbitrary shape, heterogeneity and anisotropy. The scattering condition is automatically satisfied.</td>
<td>The calculation precision is low and the size can’t be too large. The computational complexity is high and the memory consumption is large.</td>
</tr>
</tbody>
</table>
3. Analysis and discussion

Common aerosols can be divided into six categories according to their composition: dust aerosol, carbon aerosol, sulfate aerosol, nitrate aerosol, ammonium salt aerosol and sea salt aerosol. And its particle shapes are mainly spherical, ellipsoidal, cylindrical, eccentric spherical, and other complex shapes. The main components and sizes of different types of aerosols are generally different. The size range of the same aerosol is relatively small and the shapes are similar. Therefore, the methods for solving aerosol types and scattering characteristics can be roughly matched.

Cloud is a common aerosol system, but its composition is very complicated\textsuperscript{[1]}. The cloud volume of the low cloud and part of the cloud is small and the shape is approximately spherical. The particle scattering characteristics can be solved by Mie scattering theory. The particles in the upper layers of the high and middle clouds are mostly ice crystals. In the millimeter wave band, the most commonly used methods for calculating the light scattering characteristics of cirrus ice crystals are the FDTD, DDA and TMM methods. However, it is very difficult to have a unified model for calculating ice crystals’ scattering characteristics because of its complex shape.

The particle size of dust aerosol particles is mainly concentrated in the range of 0.01μm–10μm, and the dust with particle size larger than 10μm cannot be suspended in the atmosphere for a long time due to the limited transport distance. Among them, small particles with a particle size of less than 0.1μm account for more than 60%, and their ability to scatter short-wave radiation is strong. The scattering characteristics can be calculated by Mie theory, FEM or MoM method. Large particles have strong absorption capacity for long-wave radiation, and the shape is relatively uncoordinated. The PSTD method can roughly calculate the scattering characteristics, but the calculation amount is large, the memory consumption is high, and the efficiency is low; in the simulation where the precision is not too high, it can be roughly calculated using GOA and ADA.

The range of marine aerosol particle size is wide, and the water-soluble ions account for a large proportion. The peak particle size of NH\textsubscript{4}\textsuperscript{+} and SO\textsubscript{4}\textsuperscript{2−} is 0.44–1.0μm, NO\textsubscript{3}－ is 2.5–10.0μm, while Cl´
and Na$^+$ is more than 16μm$^{15}$. The particle size of these water-soluble ions is similar to wavelength, and the shape is close to a spherical shape, so it can be solved by Mie scattering theory.

Soot and volcanic ash aerosol particles are initially clusters of molecules and molecules. The majority of the particle size (83% or more) is below 0.49μm, and the average particle size is less than 0.3μm, gradually increasing with other molecules, clusters and particles. Nucleation, condensation, and agglomeration, the particle size becomes larger and larger. Moreover, the dilution ratio of air to flue gas also affects the particle size distribution of soot and volcanic ash aerosols, resulting in a relatively large particle size range, and different aggregation states result in irregular shapes. So there is no model for all particles.

4. Summary

In recent years, the research of aerosols has become a hot spot, and various remote sensing observation devices are constantly developing. The methods for studying the optical properties of aerosol particles are also innovating. In summary, the analytical method has the highest accuracy, but is limited by the special requirements of the solution, and the scope of application is the smallest; the approximation method has high computational efficiency, but the model accuracy is very low, and it is only suitable for calculations with low precision requirements; numerical method calculation The precision is high, the scope of application is wide, but the amount of calculation is large, and it develops with the advancement of computing technology. In summary, the calculation methods of various aerosol optical properties have their respective scopes of application, and they all have their own advantages and disadvantages. When studying specific problems, comprehensive consideration should be given to various factors, and appropriate methods should be selected to achieve good results.

The aerosol source composition is complex and diverse, and the shape and size are different. Although there are various theoretical models for solving the optical properties of aerosol particles, most of them are only applicable to particles of a certain scale range and special shape structure, and have not been applied to all. A high-precision unified solution model for aerosol particles. The direction of future aerosol research is to develop new observation equipment and make breakthroughs in aerosol remote sensing observations firstly. The second is to learn from the fusion signal processing technology and computational electromagnetics to promote the calculation theory and model development of aerosol optical properties, and to optimize and rationalize a more reasonable theoretical model. The third is to rely on the development of computer technology, research new algorithm programs, and simplify the model calculation process. It is foreseeable that the future calculation model of aerosol particle optical properties will inevitably develop in the direction of expanding the scope of application, improving the calculation accuracy, and accelerating the calculation speed.

References


