1 Introduction

1.1 Background

Atmospheric aerosol, as one of the most important components in the Earth-atmosphere system, is defined as solid particles (i.e., mineral dust, smoke particles, microbes, and pollen) or liquid droplets suspended in air, with diameters in the range of about 0.001 to 100 μm. The particles induced by gas-to-particle conversion processes, with diameters <0.01 μm, are known as ultrafine particles. Such small particles have been observed in bursts of very large numbers and can be converted into clusters by complex physical and chemical processes under favorable conditions. According to particle diameters, the commonly observed aerosol particles can be further classified into three categories: Aitken, accumulation, and coarse modes. The Aitken particles with diameters ranging from 0.01 to 0.1 μm act as nuclei for condensation of gaseous species under low vapor pressure, making them grow into larger particles. The accumulation-mode particles generated from aggregation of smaller particles, condensation of gases, or re-evaporation of droplets have typical diameters between 0.1 and 1.0 μm. These particles usually comprise a substantial number of soluble inorganics such as ammonium, nitrate, and sulfate, as well as carbonaceous materials. The coarse mode corresponds to particles with diameters larger than 1.0 μm, and they are primarily produced by mechanical processes and introduced directly into the atmosphere from both natural and anthropogenic sources. The diameters of coarse-mode particles and cloud droplets have certain overlapping regions; however, the diameters of coarse-mode particles are usually smaller than those of cloud droplets, and the cloud droplets easily change into rain when their diameters are large enough under favorable conditions.

The particles suspended in the atmosphere come from a variety of natural and anthropogenic sources. Those emitted from sources such as volcanic eruptions, wind-driven, or traffic-related suspensions of road, soil, and ocean, etc., are major constituents of the coarse-mode particles. The particles generated from biomass burning, incomplete combustion of fossil fuels, and industrial activities are mainly anthropogenic aerosols. Secondary particles that are mostly formed by gas-to-particle conversion in the atmosphere are the main anthropogenic aerosols that cause significant environmental issues in the urban regions. There are two main sinks of aerosol particles: dry deposition and wet deposition. Depending on aerosol properties and meteorological conditions, the lifetime of aerosol in the atmosphere ranges from hours to weeks. In addition, the particles in the stratosphere, which are mostly emitted from volcanic eruptions, can even exist for a few years and be transported all over the globe.

Aerosol particulate matters (sometimes referred to “PM” in air-quality applications) have a variety of impacts on the atmospheric environment quality. Fine-size particles (PM$_{2.5}$, whose aerodynamic diameters are <2.5 μm) are the major components of air pollutants. They have close correlations with severe health
issues including cardiovascular, lung cancer, respiratory, allergic diseases, and even enhanced mortality.\textsuperscript{5–11} In addition, the particles with small sizes produce highly effective extinction in the main spectral range of solar radiation (i.e., visible light). Therefore, PM\textsubscript{2.5} particles also cause significant reduction of visibility and have severe impacts on traffic safety and even on vegetation gross primary productivity. Moreover, aerosols exert different absorption and scattering on the electromagnetic waves in a broad spectral range from ultraviolet to near-infrared (NIR) and play a key role in the balance of global energy budgets. Due to the lack of understanding of aerosol characteristics (composition, spatial–temporal distributions, etc.), aerosols are considered to be one of the largest uncertainties in the assessment of global climate change.\textsuperscript{11} In conclusion, atmospheric aerosols have a wide variety of sources, large variations in the spatial and temporal scales, complex chemical composition, and have important impacts on the atmospheric environment and on the global climate, thus they have become a research focus in the field of geoscience.

1.2 Principles

Aerosol characteristics are complex. Due to the improvements and developments of remote sensing technology, the observable dimensions and information of atmospheric aerosol are increasing from “optics” to “microphysics,” then to “composition and species.” The main principle is based on the following context (Fig. 1).

In terms of aerosol optical observation, the aerosol optical depth (AOD) was observed initially by measuring direct sun extinction from the ground. By comparing with solar radiation outside the atmosphere, high-precision extinction measurements are obtained, which can be expressed as

$$F(\lambda) = F_0(\lambda) \exp[-m\tau(\lambda)],$$

where $F_0$ is the direct solar irradiance (W m\textsuperscript{-2}) at the top of the atmosphere (TOA) corrected by an Earth–sun distance factor, $\tau$ is the atmospheric total optical depth (by subtracting molecular contribution, AOD can be derived), $m$ is the
atmospheric mass, and \( \lambda \) is the wavelength in nanometers. Based on the spectral AOD, other aerosol parameters (e.g., Ångström exponent associated with aerosol size) can be obtained by means of semi-empirical methods and the spectral characteristics of AOD.

However, aerosol microphysical and compositional parameters can only be obtained by the inversion, which means tuning the microphysical and compositional parameters iteratively by fitting the observed radiation signals. For example, the microphysical parameters (e.g., the refractive indices) are retrieved by a commonly used optimal inversion scheme using the mathematically multi-source least squares fitting method, which allows these parameters to be simultaneously optimal. Meanwhile, as the number of retrieval parameters (e.g., about 20 particle radius bins and four wavelengths) increases, the number of observations and the measurement sensitivity become more and more important for valid inversions. At present, radiation measurements are generally collected by multi-spectral instruments, e.g., the space-borne moderate resolution imaging spectroradiometer (MODIS) sensor and the ground-based CE318 Sun–sky radiometer, as are the observations containing multi-angle information and polarized information. The inversion principles for ground-based and satellite remote sensing are quite similar; however, the contribution of a surface often dominates the observed signals in satellite inversion. Therefore, it is also required to invert the surface parameters simultaneously, such as the bidirectional reflectance distribution function.

The general structure of a standard inversion procedure is shown in Fig. 2, which can be used to retrieve aerosol optical and microphysical parameters, even for the compositional parameters. The “forward model” and “numerical inversion” are the two most complicated and elaborated modules in the algorithm. The forward model is developed to quantitatively simulate the measured atmospheric radiation reflected by the surface or atmospheric constituents with given properties. Observations are numerically inverted into aerosol size distribution,
spherical fraction, and complex refractive index using forward atmospheric radiative transfer and inversion settings. In the forward modeling of observations, the scattering phase function is one of the critical input optical parameters, which is defined as the probability distribution of photons scattered by aerosol particles in a specific direction. Since the phase function is determined by aerosol essential properties such as absorption and/or scattering, particulate size, shape, and orientation, it is related to aerosol composition. The numerical inversion module includes general mathematical operations not related to the particular physical nature of the inverted observations. The “retrieved parameters” module includes the properties of the aerosol and surface by running the numerical inversion.\textsuperscript{12}

Based on the optical and microphysical properties, aerosol composition inversion can be implemented by employing several mechanisms of aerosol composition remote sensing (Section 2) and an integrated retrieval scheme (Section 3) to obtain the optimal estimation. A simple flowchart of the inversion is shown in Fig. 3.

1.3 Historic perspective and recent advancement

Aerosol-composition remote sensing has recently been developed on the basis of the optical and microphysical parameters obtained from remote sensing observations. For optical parameters, Pouillet began to measure the solar radiation at the ground as early as 1837.\textsuperscript{13} By the 1950s, solar radiometers were designed and developed to obtain a more accurate AOD.\textsuperscript{14} From the 1980s, several regional or global networks for aerosol observation were gradually established, such as the aerosol robotic network (AERONET)\textsuperscript{15} and the Sky Radiometer Network (SKYNET).\textsuperscript{16} For satellite remote sensing, in the beginning (approximately in the 1970s), an aerosol was a by-product or an error term to be corrected from the land and ocean observations by satellite sensors. Then some specific space instruments providing global monitoring of

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**Figure 3** The flowchart of aerosol composition inversion.
aerosol properties\textsuperscript{17,18} were developed and have been running in orbit for several decades. For example, the Advanced Very High Resolution Radiometer (AVHRR) provided radiation observations in the visible and NIR wavelengths that are sensitive to aerosol properties over the ocean.\textsuperscript{19,20} MODIS has performed nearly global daily observations of atmospheric aerosols in a wide spectral range until now.\textsuperscript{21} There are several aerosol inversion algorithms for MODIS based on different principles, for example, the dark-target method for deriving aerosols over dark lands (e.g., forest) and the deep-blue method for deriving aerosols over bright surfaces (e.g., city).\textsuperscript{21–25} The multi-angle imaging spectroradiometer (MISR) measures radiance at four spectral bands (0.446, 0.558, 0.672, and 0.866 μm) and at nine distinct zenith angles, ranging from 70 deg forward to 70 deg aftward.\textsuperscript{26,27} Polarization and directionality of the Earth’s reflectance (POLDER) is a unique aerosol sensor, which is capable of measuring multi-spectral, multi-directional, and polarized radiance.\textsuperscript{28} Because of these advantages, POLDER data are better suited to determine the size and shape of particles over the ocean,\textsuperscript{29} which can be used in the retrieval of fine-mode AOD. However, at present, the only aerosol parameter over land obtained from satellite remote sensing with a high accuracy is AOD. Other advanced satellite aerosol products are under test and are expected to be released during the next several years. In addition, a Lidar system can provide vertical information about aerosols, which is another remote-sensing approach but with active measurements. The related Lidar systems include ground-based networks such as the European Aerosol Research Lidar Network (EARLINET),\textsuperscript{30} the Lidar Network in Latin America (ALiNet),\textsuperscript{31} the Commonwealth of Independent States Lidar Network (CIS-LiNet),\textsuperscript{32} and the GAW Aerosol Lidar Observation Network (GALION),\textsuperscript{33} and space-borne satellites, e.g., the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO).\textsuperscript{34}

In the past four decades, numerous inversion methods have been developed to interpret the measured radiative characteristics of atmospheric aerosols. For example, the algorithms of King et al.,\textsuperscript{35} Nakajima et al.,\textsuperscript{36,37} and Wang and Gordon\textsuperscript{38} proposed for obtaining aerosol optical and microphysical properties from atmospheric radiances are well-established. The statistical method developed by Dubovik and King\textsuperscript{39} retrieved an extended set of aerosol parameters (including aerosol-particle size distribution and complex refractive index) by simultaneously inverting all available multi-angular and multi-spectral measurements of atmospheric radiances to maximize the retrieved aerosol information, which is now the standard method of ground-based remote sensing. At the same time, Dubovik et al.\textsuperscript{12} also proposed a generalized retrieval of aerosol and surface properties (GRASP) inversion framework, which has been reprocessed based on POLDER historical data. GRASP is a rigorous and versatile algorithm divided into several interacting but rather independent modules to enhance its flexibility (Fig. 2). A statistically optimized fitting of observations based on the multi-term least squares method is employed to combine the advantages of a variety of approaches and provide transparency and flexibility in developing algorithms that invert passive or active observations and derive several groups of unknown parameters.\textsuperscript{30}
Similar to the development process of remote sensing of optical and micro-physical parameters, the aerosol composition inversion was first proposed by researchers using ground-based remote sensing observations and then tested with satellite observations. Although there are continuous observations from satellite remote sensing for more than 20 years with global coverage, the obtained parameters are mainly optical. Therefore, especially over land, the satellite-derived parameters reflecting aerosol “composition and component” are far from mature, which is mainly attributed to the surface information contained in the satellite observations and also to limited sensitivity. However, with the development of multi-dimensional (multi-spectral, multi-angle, and polarization) sensors, such as POLDER on board the Polarization and Anisotropy of Reflectances for Atmospheric Science coupled with Observations from a Lidar (PARASOL) and directional polarimetric camera (DPC) onboard the GF-5 satellite, the retrieval of aerosol composition from satellite observation has basically reached its singular point. Therefore, it is necessary to summarize the principles, current situations, mechanisms, methods, and applications, which are the main contents of this Spotlight, to prepare for the following advancement (Table 1).

2 Mechanism of Aerosol-Composition Remote Sensing

2.1 Optical absorbing components

There are four major kinds of absorbing aerosol species in Earth’s atmosphere, including black carbon (BC), brown carbon (BrC), hematite, and goethite. BC and BrC are usually emitted by similar combustion sources and generally coexist in aerosol mixtures. Hematite and goethite are iron oxides that are different forms of “free” iron and typically appear together as well. In order to distinguish the discrepant contributions of these absorbing species, aerosol light absorption properties, such as at the imaginary part of the refractive index and single-scattering albedo (SSA), are utilized.

2.1.1 Black carbon

Chain aggregates of BC (Fig. 4) containing soluble compounds can collapse to form sphere-like structures when subjected to increasing relative humidity (RH). They are often encapsulated in inorganic salts and organic matters (OMs). The BC is a significant absorbing component that is wavelength independent and has high real and imaginary parts of the refractive index (RRI and IRI). By calculating an equivalent refractive index for the aerosol mixture, which is described as very small carbonaceous particles spreading throughout a host, we can use Mie theory to estimate the optical properties of the BC component.

2.1.2 Brown carbon

BrC is an organic mixture of carbonaceous materials. A distinct property of BrC is that its imaginary part of the refractive index is substantial at ultraviolet (UV)