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Using a new aerosol relative optical thickness concept to identify aerosol particle species

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ABSTRACT

We developed an aerosol relative optical thickness concept and then established an effective aerosol particle recognition model by analyzing variations in aerosol optical thicknesses in Beijing between 2001 and 2006. The accuracy of the model was verified using inverse calculations. The aerosol particle types and size distributions were assessed for several typical atmospheric phenomena, and the characteristic relative optical thicknesses for several typical aerosols were identified. Finally, we analyzed annual variations in the aerosol particle species in several eastern Asian cities using the model.

1. Introduction

Atmospheric aerosol is a multiphase system (Ziemann, 2010), involving solids, liquids, and gases, with diameters between 0.001 and 10 μm. Aerosols can be of natural or anthropogenic origins, and affect the Earth's radiation budget (Pope et al., 2012) through direct and indirect effects, playing an important role in global climate systems. The direct effect on the radiation budget is that aerosol particles scatter and absorb solar radiation and surface infrared radiation, and the indirect effect is that aerosols act as cloud condensation and ice nuclei (Wang, 2013; Koren et al., 2004; Ghan et al., 2013). Atmospheric CO_2 concentrations have increased by 25% in the past 100 years because of fossil fuel and biomass combustion, and the terrestrial surface temperature has risen by about 1 K, globally (Meehl, 1996; Le Treut, 1998). According to the fourth assessment report of the Intergovernmental Panel on Climate Change (Le Treut et al., 2007), about the same amounts of global average radiative forcing are caused by aerosols and greenhouse gases, but they have opposing effects, the radiative forcing value can reach -1.2 W/m^2. Aerosols have complex origins and the phrase time intervals in the atmosphere, and their characteristics clearly vary over time and space. The influence of human activities on the global environment is increasing and research into the effects of aerosols on the atmospheric environment is a growing field (Kirkby et al., 2011). Atmospheric aerosols exert an important influence on climate. The impacts arising from industrial emission, vehicle exhaust, and other factors adversely affect human health. It is, therefore, important to identify and predict concentrations, species and particle size of aerosols, and thus to investigate the formation mechanism of the aerosol particles.

The aerosol particle observation includes direct and indirect observations. The direct observation is to assay, analyze and count the aerosol particles by the way of collecting and sampling. The indirect observation is to deduce the concentrations, species, particle size and other information of the aerosols by observing their physical and chemical characteristics. In recent years, two main observational technologies, satellite remote sensing (Feng and Christopher, 2013; Xu et al., 2012) and ground-based

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observation, have been developed to measure the radiative characteristics of atmospheric aerosols. The AERONET (aerosol robotic network), which was initiated and supported by the United States National Aeronautics and Space Administration, has been used for ground-based remote sensing, and has been accepted as being the most advanced system in the world (Radhi et al., 2010). This network includes more than 500 observation stations distributed around the world, and its main purpose is to obtain regionally representative parameters for aerosol optical characteristics using ground-based CIMEL sun photometers, on a global scale. The observational data from this network can be used to verify the aerosol optical characteristics obtained from satellite remote-sensing observations. The AERONET stations are mainly in cities in Europe, the United States, and eastern Asia. Unlike the satellite remote-sensing observations, the AERONET data are temporally discontinuous, that is, the time intervals between measurements are not consistent between stations, and not even for the same station. The global observation stations of AERONET have collected a large number of observational data with wide applications, such as the comparison with satellite data and the investigation of ground aerosol characteristics. But the utilization of these data is not sufficient. So effectively utilizing these data is an important problem needed to be solved.

The radiative characteristics of atmospheric aerosols vary widely because of a number of factors, including the types of aerosol-forming human activities taking place (e.g., vehicle exhausts, heating plants), seasonal variations (e.g., spring sand storms in eastern Asia), and differences specific to a location, and the cycles for these factors are different. For instance, emissions from vehicle exhausts follow a daily cycle, greatly influencing atmospheric aerosols in the daytime but having little influence at night. Sand storms only occur in the spring, and each storm lasts for a short time. Aerosol properties are also different in maritime and inland cities. It is very important, but difficult, to be able to characterize the ranges of aerosol particles present from the large quantities of aerosol observation data that are collected.

To attempt to solve this problem, we analyzed the observational aerosol data from eastern Asia, and classified the data to identify different aerosol types. We distinguished aerosols caused by dust storms from other aerosols using the aerosol optical thicknesses (AOTs) in spring. We aimed to identify the aerosol particle species more specifically than the currently available tools allowed, so we developed a new aerosol relative optical thickness (AROT) concept. Using the aerosols measured in Beijing between 2001 and 2006 as an example, we verified the AROT concept using a multi-log-normal distribution model based on a stochastic particle swarm optimization (SPSO) algorithm. Finally, we analyzed the AROT variations observed in several cities in eastern Asia, and identified the variations in the aerosol particle species that caused them.

2. Identifying aerosol particles using spectral optical thickness

We first studied the dust-dominated spring weather in Beijing. Fig. 1 shows the AOT observation data for 2001–2006, and it can be seen that the peak AOT values were higher than 2.5 in the springs of 2001, 2003, and 2004, when the atmospheric aerosol particle concentrations were clearly higher than at other times (that is, the air was heavily polluted). The AOTs in Beijing fluctuate dramatically because of automotive exhaust emissions, plant activities, and other emissions, and can reach a peak of about 2.5 or can fall to less than 1. However, the patterns for different AOTs are distinct. For example, it can be seen from the circled observational data in Fig. 1c that AOTs at a wavelength of 0.440 μm can reach values higher than 3.8, and follow a very different pattern from the patterns seen at other wavelengths. The AOT is about 2.6 at a wavelength of 0.694 μm and is only 1 at the lowest wavelength, 1.02 μm. The AOTs decrease with increasing wavelength. In contrast to the phenomenon described above, the spectral AOTs observed in 2001 (Fig. 1a) were similar to each other. The distinct AOT values seen in Fig. 1a–f may have been caused by variations in the aerosol types and particle size distributions, amongst other factors, which cause the characteristics of the aerosol particles to vary.

Fig. 2 shows the changes in the aerosol optical characteristics at different times in 2001 and 2003, and clearly shows that there were significant variations in the aerosol properties in Beijing. As can be seen in Fig. 2a, the spectral AOTs varied only slightly in 2001, The AOTs at the four wavelengths illustrated all being between 2 and 2.5. According to our previous analyses (Yuan et al., 2011), the aerosol particles were mainly concentrated in the coarse mode, that is, with an average particle size of about 1 μm. This agrees well with what would be expected of dust aerosols, although such particle species are not common in urban aerosols. The observational data from 2003 showed great fluctuations, clearly indicating that the particle components and size distributions changed dramatically. Several aerosol types may be present at any time. If we can identify the aerosol types and particle size distributions, we can then estimate the amount of dust aerosol that is present in the atmosphere.

We analyzed the aerosol variations over time, as is shown in Fig. 2, assuming that the particle species were dust and that the spectral complex refractive indices were shown in Table 1. The aerosol particle size distributions had log-normal distributions. Three modes were considered in the distribution analyses, Aitken mode ($\overline{D_1} = 0.05\mu m$), accumulation mode ($\overline{D_2} = 0.04\mu m$), and coarse mode ($\overline{D_3} = 1.0\mu m$), all with the same standard deviation, $\beta_1 = \beta_2 = \beta_3 = 0.307$. The number densities, N_i, of the particles were calculated using the retrieval model, and more details can be found in our previous publication. The convergence accuracy was set as 10^{-10}, and the maximum number of iterations was 3000.

The inversion results for the four selected 2001 samples were good. The fitness values for the three calculated samples were below 10^{-10}, with generation numbers of less than 1000, and the calculated sample on April 8, 2001 was very accurate at a generation number of 3000. We found that the estimated aerosol properties for 2001, using the above method, were accurate, and, therefore, that the method was suitable for setting the particle size distribution, the complex refractive index, and other parameters. However, the retrieved results for the aerosol observation samples from 2003 were not acceptable. Except for the data for April 13, 2003, the fitness values for the samples were only about 0.1 at a generation number of 3000. The variations in each spectral optical thickness were similar in the April 13, 2003 observations, as shown in Fig. 2b, which is consistent with the 2001 results. This illustrates that the dust aerosol assumption was applicable to the 2001 and April 13, 2003 samples but not to the samples for the other three dates,