Effective absorption for Black Carbon-NaCl internal mixed aerosols

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Abstract: Effective absorption of internal mixed aerosol has severe negative effects on laser transmission in atmosphere. The effective absorption coefficients of homogeneous sphere model, BC-core model and NaCl-core model aerosols, with the same density and specific heat of particle, were discussed by Mie theory. Results show that effective absorption is associated with comprising method. The BC-core sphere particles mainly show superior effective absorption at short wavelength as visible light and infrared light, homogeneous sphere particles mainly super at mid-infrared wavelength and far-infrared wavelength after 100 μs, while NaCl-core particles mainly show larger effective absorption coefficient at far-infrared wavelength at beginning of heating procedure.

Key words: aerosol; effective absorption; internal-mixed; Mie scattering

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黑碳-氯化钠内混合气溶胶粒子有效吸收研究

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摘 要: 内混合气溶胶粒子的有效吸收对激光大气传输有着不利影响。以黑碳和氯化钠两种典型成分为例,计算了密度和比热相同情况下,均匀球和分层球模型粒子的有效吸收系数。研究结果表明:有效吸收系数大小与粒子组分混合方式有关。在粒子吸收热量加热大气过程中,黑碳为核的粒子在可见光和近红外波段有效吸收系数更大;均匀球粒子在中红外波段和远红外波段(100 μs 后)有效吸收系数更大,而氯化钠为核的粒子在远红外波段(前 100 μs)有效吸收系数最大。

关键词: 气溶胶; 有效吸收; 内混合; Mie 散射

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0 Introduction

Aerosol has become a ubiquitous topic in fundamental research, giving access to its various physical and chemical properties in the field of atmospheric radiation transfer and laser transmission. Researchers have paid great attentions on aerosol optical characteristics due to its negative effect on laser propagation in atmosphere, and influence on so many aspects such as lidar, laser communication and laser guided weapon[1-4]. The diversity of aerosol types results in a complex mixture of aerosol components that reside in the atmosphere. Differences of components and structure modes will affect the optical characteristics of the aerosols. Internally mixed primary aerosols are produced at the source if more than one component is involved in the creation of the aerosol. This can be expected in the combustion of complex mixtures of materials such as fossil fuels and biomass. It has been reported that wind-generated sea-salt aerosols created by the bursting of air bubbles which ejects water droplets that can contain organic material from the ocean surface which will mix with the sea salts as the droplet evaporates [5-6]. Even if the internal composition of a given aerosol is known, there are still uncertainties related to the distribution of the various components within the aerosol. If the internal mixture formed because of coagulation of sulfate aerosols onto a larger dust particle, then the appropriate model would be a coated aerosol with the dust at the core and the sulfate comprising the shell. On the other hand, if the internal mixtures are formed from the evaporation of cloud droplets that grew by coalescence, then a well-mixed matrix of substances would be a better model. The location of the various components has consequences on computing the optical properties[7-10].

When a high-intensity laser beam propagates through the atmosphere containing aerosols, a portion of the beam energy is absorbed by the aerosol particles. Research es have shown that the absorption peculiarities are different for aerosols made of different components. However, aerosol effective absorption coefficient is just a part of absorption efficient, corresponds to the part of aerosol absorption that heats up the air only. This energy heats the particle directly, and after a short time, the air is heated by thermal conduction from the particles. Unlike the case of molecular absorption, aerosol effective absorption is time dependent, owning to the small but not vanishing heat capacity of aerosol particles. Differences in components affect effective absorption for single component particles. Comparing with carbon dust, water droplet has smaller effective absorption coefficient ($\alpha_{eff}(t)$) with small absorbency and large heat capacity [11]. Other Research also shows that the effective absorption coefficient is obvious for the short wavelength for aerosols made of black carbon and water by internal-mixing. The initial growth of $\alpha_{eff}(t)$ in the 1-100 μs period is much more rapid than that in the 100 μs-1 s period. When the system becomes balanced, $\alpha_{eff}(t)$ becomes constant with the same value as the absorption coefficient [12]. Research also shown for uniformly mixed hygroscopic aerosol consists with either sodium chloride and water or ammonium sulfate and water, the relative error of the coefficient, under the case of considering the absorption by the particles themselves and the case of ignoring, are 234.7% 和 255.2% at the time 1s, respectively[13].

For internal-mixed spherical aerosols made of carbonaceous material and sea-salt, there are two typical kinds of symmetrical sphere models: homogeneous sphere model and core-shell sphere model. For the later model, two kinds of particles are calculated by changing comprising method. One of particular interests is the treatment of black carbon since its high absorption coefficient makes it particularly sensitive to mixing assumptions, so black carbon (BC) and sodium chloride (NaCl) are used to study the effective absorption of aerosol made of carbonaceous material and sea-salt [14]. In the paper, the volume ratio of two compositions is 1:1, so that the density and specific heat of different models is exactly same. The results shed new light on the transmission of high power laser in atmosphere, and in favor of the research of formation mechanism for thermal blooming caused by aerosol.

1 Theoretical model

The effective absorption for a given aerosol can be defined as[11]

$$\alpha_{\text{eff}}(t)=\int \pi r^2 Q_{\text{abs}}(r)n(r)\{1-\exp[-t/\tau(r)]\}\mathrm{d}r \tag{1}$$

where r is radius of the particle, and $Q_{\text{abs}}(r)$ is absorption efficiency factor. $n(r)$ represents the number of aerosol particles per unit volume with a radius range from r to $r+\mathrm{d}r$, and $\tau(r)$ is the time to reach the eventual temperature of the particle above the ambient temperature,

$$\tau(r)=r^2\rho c/3k \tag{2}$$

where ρ and c are the density, and specific heat of particle, respectively, $k=2.6\times10^{-2}$J/(s·m·K), which represents thermal of air.

For black carbon (BC) and NaCl internal mixed aerosol particles, two typical kinds of sphere models are used: homogeneous sphere model and core-shell model. Defining BC and NaCl with a volume ratio 1:1, ignoring the volume change caused by mixture, so that no matter what the distributions of the components, particles have several same physical properties, such as density and specific heat.

For homogeneous sphere model, the refraction index and heat absorbability of particle can be written as:

$$m=m_2+(m_1-m_2)\ (V_1-V_2) \tag{3}$$

$$c\rho V\Delta t=c_1\rho_1V_1\Delta t+c_2\rho_2V_2\Delta t \tag{4}$$

where m and V represent refraction index and volume, respectively. The subscript 1 and 2 represent two components of the particle and non-subscript represents the parameter of the whole particle.

For particle with a component volume ratio of 1:1,

$$V_1=V_2$$

from Eqs.(3)、(4), we can get

$$m=(m_1+m_2)/2 \tag{5}$$

$$c\rho=(c_1\rho_1+c_2\rho_2)/2 \tag{6}$$

For core-shell model, there are two kinds of particles, BC-core with NaCl-shell (i.e. BC-core) and NaCl-core with BC-shell (i.e. NaCl-core). The absorption contains two segments, the core absorption and the shell absorption,

From Eq.(4), it can be found that

$$c\rho=\frac{c_b\rho_b(b^3-a^3)+c_a\rho_a a^3}{b^3} \tag{7}$$

where a and b are the radius of the core and the whole particle respectively. ρ_a, c_a, ρ_b and c_b are the density and specific heat of core and shell, respectively.

For particle with a component volume ratio 1:1,

$$b^3/a^3=2 \tag{8}$$

Inserting Eq.(8) to (7), we can find

$$c\rho=\frac{(c_a\rho_a+c_b\rho_b)}{2} \tag{9}$$

For particle made of black carbon and NaCl,

$$c\rho=\frac{(c_{\text{NaCl}}+\rho_{\text{NaC}l}+c_{\text{BC}}\rho_{\text{BC}})}{2} \tag{10}$$

whether it is homogeneous sphere model or core-shell model.

Inserting Eq.(10) into Eq.(2), it gets

$$\tau(r)=r^2(c_{\text{NaCl}}+\rho_{\text{NaCl}}+c_{\text{BC}}\rho_{\text{BC}})/6k \tag{11}$$

The parameters used for specific heat and density of NaCl and BC are

$$c_{\text{NaCl}}=2.329\times10^3\text{J/(kg·K)}$$

$$\rho_{\text{NaCl}}=2.165\times10^3\text{J/(kg·K)}$$

$$c_{\text{BC}}=0.714\times10^3\text{J/(kg·K)}$$

$$\rho_{BC}=2.25\times10^3\text{J/(kg·K)}$$

The complex indices of refraction for different wavelengths of incident light are shown in Tab.1[15-20].

The size distribution can be given by Jungle′s formula[12,21].

Tab.1 Complex indices of refraction for black carbon and NaCl

Wavelength/μm	Black carbon	Black carbon
0.55	m_{BC}=1.75−0.44i	m_{BNaCl}=1.547−6.8×10^{-11}i
1.06	m_{BC}=1.75−0.44i	m_{BNaCl}=1.531−3.5×10^{-10}i
3.75	m_{BC}=1.90−0.57i	m_{BNaCl}=1.522−1.8×10^{-9}i
10.6	m_{BC}=2.20−0.90i	m_{BNaCl}=1.491−8.0×10^{-8}i

$$n=\begin{cases}2\times10^{-12}\times r^{4} & \text{for } 2\times10^{-12}\ \mu\text{m}<r<30\ \mu\text{m}\\ 0 & \text{elsewhere}\end{cases} \qquad (12)$$

From Mie theory [22-24], we can get the absorption efficiency factor Q_{abs}. Inserting Eqs.(11) and (12) into Eq.(1), the effective absorption coefficient of different models can be obtained.

2 Results and discussions

Figure 1 shows that the $\alpha_{eff}(t)$ gradually increases with time until it becomes saturation and reaches a stable value which equals to the absorption coefficient. It means at this time point, the energy absorbed by particles is contra-balanced with the energy used to heat the air around them. The figure also demonstrates that increasing rate declines with time. During the procedure, the effective absorption is more remarkable at short wavelength, just as the absorption coefficient. If the laser powers are the same for 0.55 μm, 1.06 μm, 3.75 μm and 10.6 μm, the thermal blooming more likely occurs at short incident wavelength.

The values of $\alpha_{eff}(t)$ are quite different from each other although they have the similar variation profiles. For the incident wavelength of 0.55 μm and 1.06 μm, the order of $\alpha_{eff}(t)$ value is just the same as that of $\alpha_{eff}(t)$ for the three kinds of particles. For 0.55 μm, the α_{eff} (t) of particle with BC-core and NaCl-shell (i.e. BC-core) is more prominent, homogeneous sphere model comes secondly, and that of particle with NaCl-core and BC-shell (i.e. NaCl-core) NaCl-core is the smallest during the procedure of particle heating the air, as shown in Fig.1(a). For 1.06 μm, the $\alpha_{eff}(t)$ of NaCl-core particle is still the smallest during the

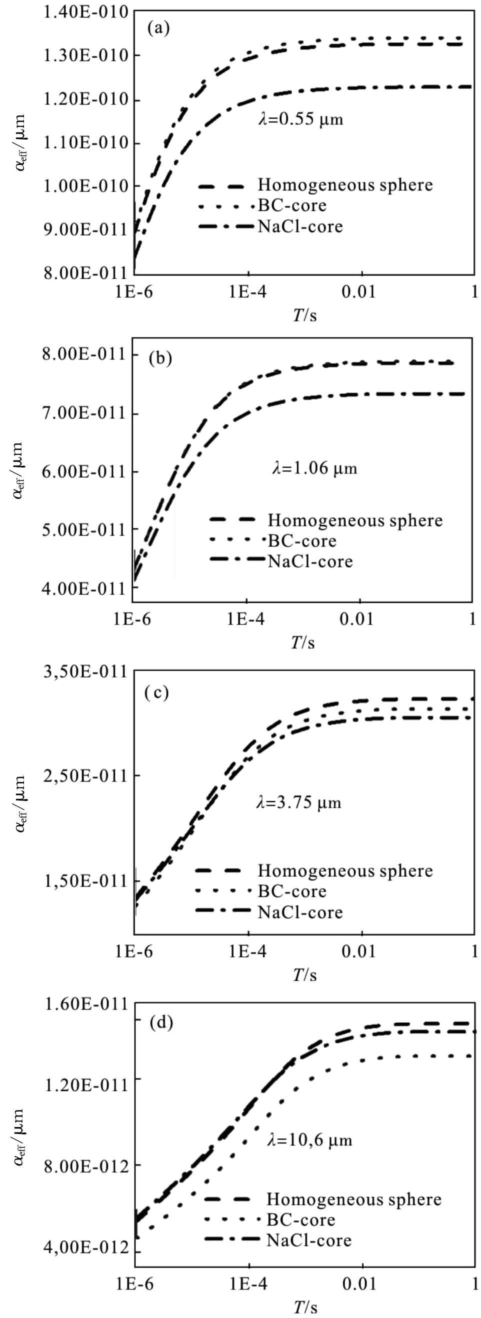

Fig.1 Effective coefficient variation of BC-core sphere model, NaCl-core sphere model and homogeneous sphere model at incident wavelength of (a) 0.55 μm, (b) 1.06 μm, (c) 3.75 μm, and (d) 10.6 μm

procedure. The curve of homogeneous sphere and BC-core particles are almost coincident, just as their values of $\alpha_{eff}(t)$. It indicates that absorption capacity is a significant factor for particles on the dominance of effective absorption coefficient.

At the incident wavelength of 3.75 μm, homogeneous sphere particle takes the place of BC−core particle and becomes dominance at $\alpha_{eff}(t)$. Besides, it has a highest $\alpha_{eff}(t)$ along the heating process. A cross point can be observed at about 30 μs, implying the $\alpha_{eff}(t)$ of BC−core and NaCl−core particle reaches a same value, as shown in Fig.1(c). At the beginning, the effective absorption coefficient of NaCl−core particle is superior, even though its absorption coefficient aerosol is smaller. After the crossing time, the BC−core particle overtake NaCl−core particle at the value of $\alpha_{eff}(t)$, the advantage increases with time. It indicates that the $\alpha_{eff}(t)$ is not only depend on absorption coefficient, but also relevant to heat conduction between particles and the ambient air.

For 10.6 μm, as shown in Fig.1 (d), the $\alpha_{eff}(t)$ value relation of two core−shell particle is obvious on the contrary to that of 0.55 μm. This exchange in dominate position is quite similar with their absorption coefficient[8]. The $\alpha_{eff}(t)$ of NaCl−core particle is even higher than that of homogeneous particles before 100 μs.

The order of $\alpha_{eff}(t)$ for three particle models changes with wavelength. If the laser wavelength is longer than the size of particle wave optics implies that BC−core particle has a strong absorption; on the contrary, if the laser wavelength is shorter than the size of particles, geometric optics implies that it has a weak absorption compared with NaCl−core aerosol[23].

3 Conclusion

The effective absorptions have been investigated numerically with homogeneous sphere model, NaCl−core sphere model and BC−core sphere model consist of black carbon (BC) and sodium chloride (NaCl) with the same volume.

The effective absorption coefficient ($\alpha_{eff}(t)$) gradually increases with time until it becomes saturation and reaches a stable value which equals to the absorption coefficient while the energy absorbed by particles is contra−balanced with the energy used to heat the air around them. During the absorption procedure, the effective absorption is more remarkable at short wavelength, just as the absorption coefficient.

The BC−core sphere particles mainly show superior effective absorption at short wavelength as visible light (0.55 μm) and infrared light (1.06 μm), homogeneous sphere particles mainly super at mid−infrared wavelength (3.75 μm) and far−infrared wavelength (10.6 μm) after 100 μs, while NaCl−core particles mainly show larger effective absorption coefficient at far−infrared wavelength (10.6 μm) at beginning of heating procedure.

This research is helpful for obtaining a clear understanding of the formation mechanism of aerosols and their properties, which have distinctive influence on thermal blooming effect. It also provides good suggestion for parameter optimum in the laser system design. The results can be used to evaluate beam quality deterioration caused by aerosols when high energy laser propagation in atmosphere, especially for long laser pulse. Based on this finding, further work about the thermal distortion and beam expansion caused by aerosols in atmosphere is needed.

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