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Calculation of optical properties of light-absorbing carbon with weakly absorbing coating: A model with tunable transition from film-coating to spherical-shell coating

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ABSTRACT

Optical properties of particles consisting of light-absorbing carbon (or soot) and a weakly absorbing coating material are computed at a wavelength of 355 nm and 532 nm. A morphological particle model is used, in which small amounts of coating are applied as a thin film to the surface of the aggregate, while heavily coated aggregates are enclosed in a spherical shell. As the amount of coating material is increased, a gradual transition from film-coating to spherical-shell coating is accounted for. The speed of this transition can be varied by specifying a single parameter. Two different choices of this parameter, corresponding to a slow and a rapid transition from film-coating to spherical-shell coating, respectively, are investigated. For low soot volume fractions the impact of this transition on the linear depolarisation ratio δ_l is most pronounced. The model that describes a rapid transition to a spherical coating yields results for δ_l that are more consistent with existing lidar field measurements than the slow-transition model. At 532 nm the relative uncertainty in modelled δ_l for a rapid transition values due to uncertainties in the aggregate's geometry and chemical composition are estimated to range from 109 to 243%, depending on the soot volume fraction. At 355 nm the relative uncertainties were estimated to range from 90.9 to 200%.

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1. Introduction

Particles consisting of light-absorbing carbon or soot are among the atmospheric aerosol particles with the largest impact on the Earth's climate. They can influence the climate system through their role in cloud formation processes, by reducing the albedo of snow and ice in polar and mountainous regions, or by direct absorption of solar radiation [1,2]. Also, soot particles have an adverse impact on air quality and human health [3]. Observations of soot particles by remote sensing techniques play an important role in monitoring sources, transport pathways, and deposition of soot particles in the atmosphere. These observations are also essential for constraining and improving aerosol transport and air quality forecasting models. The interpretation of remote sensing observations as well as predictions of the climate impact of soot particles relies on a thorough understanding of the particles' optical properties.

Soot particles themselves consist of highly absorbing carbonaceous material. Aged atmospheric soot particles are commonly coated by weakly absorbing material, which can complicate the modelling of optical properties of soot particles [4]. Both images of atmospheric soot particles and chemical analyses often indicate a thick coating and hence a low soot volume fraction [5–8]. In [8] 50% of freshly emitted soot particles were reported to be heavily coated, thus indicating a fast coating process in the atmosphere. As shown in [7] the coating itself may consist of different materials.

Various models have been employed to investigate the impact of the particles' morphological features on radiative and optical properties. The extent to which morphological features of coated soot particles need to be resolved in models depends on the intended application. To account for the coating different particle models have been used. The climate relevant impact on the broadband solar radiative flux can be investigated by assuming a spherical core-shell or the recently introduced core grey shell model [9]. Using morphologically more complex particle models requires more computational efforts. The impact of the use of simplified particle models in climate models can be quantified by compar-

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ing the results obtained for simplified models to those obtained for more complex models (e.g. [10]).

Prior work on the uncertainty introduced into climate models by assuming strongly simplified shapes of soot particles focused on radiative properties, such as the mass absorption cross section (e.g. [11]) or the scattering and absorption cross sections [12,13], as well as on the single scattering albedo and asymmetry parameter. The closed cell or concentric core-shell monomer model employed by [13–15] uses aggregates where each individual monomer is coated by a spherical shell. Other approaches apply the coating onto the aggregate of contacting soot monomers. Two other approaches in which the coating follows the shape of the aggregate were used in [13]. The first approach defines a fixed coating thickness. A coating layer with this thickness is then added to the aggregate. The other approach adds small coating volumes at the edges of the soot aggregate or the coating until a prescribed volume fraction is reached. In another study coating was added by first filling voids in branches with more densely packed monomers; then the coating is continued in descending order of the density of monomer packing [12]. Partial soot inclusions in spherical coating shells were investigated in [12,16]. Both the model which filled voids in branches and the partial soot inclusion model discussed in [12] served as a references for comparison with simpler core-shell and homogeneous sphere models. However there were differences between the two morphologically more complex models. The scattering cross section is larger if the surface of coated soot increases. In [16] the impact of monomer surfaces intersecting with the coating surface was investigated. The difference between the two models was less than 5% for optical cross sections, asymmetry factor and single scattering albedo; therefore, this morphological feature may be ignored.

A numerical investigation presented in [17] was carried out to evaluate the possibility of determining the primary particle size of bare soot aggregates by measuring the depolarisation of light. In that numerical investigation it was found that refractive index, the aggregate shape and connections of the monomers making up the aggregate have a strong impact on the depolarisation.

Compared to climate models, remote sensing applications may require morphologically more complex models. One quantity frequently used in lidar remote sensing is the linear depolarisation ratio, δ_l. This quantity is a key element in classifying aerosol species using lidar techniques. The aerosol classification schemes of the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) [18] and for the planned Earth Clouds, Aerosols and Radiation Explorer (EarthCARE) [19,20] rely on measurements of δ_l. Depolarisation is highly sensitive to morphology, and even to particle inhomogeneity [21]. It is therefore expected that a correct description of depolarisation requires significantly more realistic model particles than radiative forcing computations in climate models.

Images of coated soot particles presented by [6,22] suggest that the coating in case of low soot volume fraction tends to form a spherical or almost spherical shape, which encapsulates parts of the soot aggregate, while parts of the aggregate are sticking out of the coating. A corresponding model has been considered in, e.g., [9,16]. However, this model is not expected to be very realistic for high soot volume fractions (i.e., thin coatings). For those the coating forms a thin film that closely follows the shape of the aggregate, similar to the model in [13]. It is plausible to assume that there should be a smooth transition from the nonspherical to the spherical coating, which would take place at intermediate soot volume fractions. Also, as soot particles age in the atmosphere, their fractal dimension tends to increase, resulting in a collapse and compaction of the aggregate structure [4,23–25]. In [8] the particles with the lowest soot volume fraction were reported to have the highest fractal dimension, i.e. being the most compact particles. Particles with higher soot volume fractions were reported to have lower fractal dimension.

Thus, most coating models considered earlier can be expected to be realistic either for low or for high soot volume fractions. A recent study presented a first attempt to cover the entire range of volume fractions by devising a coated aggregate model that accounts for the transition from thinly coating films to spherical coatings [26]. However, the fractal dimension has been assumed to be constant, i.e., the transition from lacy to more compact soot aggregates during the coating process has been neglected. The computational results were compared to those obtained with the closed cell model. The cross sections of both models were in relatively good agreement for high soot volume fractions (i.e., thin coatings). However, the depolarisation ratios were found to differ between the two models. The depolarisation ratios of soot particles obtained with the coated aggregate model were more consistent with field observations than those obtained in the closed cell model. However, there seems to be a certain risk that the coated aggregate model overestimates depolarisation at low volume fractions and for large particle sizes. This suggests that the model particles proposed in [26] might be too nonspherical for low soot volume fractions.

To lower the depolarisation by soot particles one could make the transition from thinly-coating films to spherical coatings more rapid, because this will make particles with low soot volume fractions more spherical. This can be achieved by (i) reducing the radius of the critical sphere defined in [26] that marks the transition from film-coating to spherical coating; and (ii) assuming that the fractal dimension of the aggregate increases with decreasing soot volume fraction (i.e., increased coating thickness).

We hypothesize that the *speed of transition* from film-coating to spherically encapsulated soot aggregates is an essential morphological parameter to which the linear depolarisation ratio is highly sensitive. By a *high* speed of transition, we mean that the coating becomes spherical at relatively small amounts of coating material (i.e, at relatively high soot volume fractions); while a *low* speed of transition requires higher amounts of coating material before the shell becomes spherical.

We will test our hypothesis by formulating a particle model in which the speed of transition can be continuously adjusted by tuning a single parameter. We will then compare two choices of this parameter, which simulate a slow and a rapid transition, respectively, from film-coating to spherical coating as more liquid-phase material is added to the soot aggregate. The computational results for the linear depolarisation ratio are gauged against published observations from field measurements.

A more detailed description of the particle model, as well as a description of the computational methods are given in Section 2. The results are presented and discussed in Sections 3 and 4, respectively. Concluding remarks are given in Section 5.

2. Methods

2.1. Particle models

Atmospheric soot aerosol particles can be approximated as aggregates consisting of spherical monomers following the scaling relation [27]:

$$N = k_0\left(\frac{R_g}{a}\right)^{D_f} \tag{1}$$

In Eq. (1) N denotes the number of monomers, a the monomer radius, k_0 is the fractal prefactor and D_f the fractal dimension. The fractal dimension takes values between 1 (linear aligned monomers) and 3 (spheres). The radius of gyration R_g is given by

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