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Modeling the optical properties of combustion-generated fractal aggregates

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HIGHLIGHTS

- Aerosol properties measured for both flaming and non-flaming combustion.
- Properties used to test model predictions.
- Parameters necessary for model predictions to be accurate are obtained and discussed.

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ABSTRACT

Combustion-generated carbonaceous aerosols are generally in the form of fractal aggregates (FA's) with shapes that vary from long chain-like structures to much more compact, almost spherical structures, depending upon the mode or stoichiometry of the combustion process. Typically, as combustion moves from fuel-lean to fuel-rich, aggregate morphologies change from the former to the latter. Accompanying this change in morphology is a change in the chemistry of the aggregates as the percent of carbon in the aggregates also decreases. These combined changes produce radically different scattering and absorption signatures that define their radiative transfer properties. To improve our ability to predict how these optical properties change, experiments were conducted to measure both the physical and optical properties of these aggregates for both flaming and non-flaming modes of combustion. Using the aggregate property data from these experiments, numerical calculations were then performed using both the discrete dipole approximation (DDA) and the Rayleigh–Debye–Gans (RDG) approximation to generate their characteristic scattering and absorption signatures. This paper presents the experimental results, the comparison of the modeling results with the experimental results and discusses those parameters most important to obtain agreement between the modeling and the experiments.

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1. Introduction

Combustion processes generate aerosols as a natural by-product with varying sizes, morphologies, and chemical compositions that depend not only upon the combustible solid, liquid or gas but also upon the mode of combustion, such as flaming or non-flaming, or the stoichiometry of the combustion process, ranging from oxygen-rich to fuel-rich. These aerosols are generally in the shape of fractals or fractal-like aggregates, with individual aggregates containing 10–100's of smaller primary particles connected in some random fashion. In general, for over-ventilated, or oxygen-rich, combustion, these aggregates appear as elongated, chain-like structures with characteristic fractal, or Hausdorff, dimensions, D_f, in the range of 1.6–1.9. For aggregates that are produced from non-flaming or fuel-rich combustion, the structure is more compact and clumped with fractal dimensions typically in the range of 2.1–2.3. Knowledge of the structure, the size and the chemistry of these aggregates once expelled into the atmosphere is important in the subsequent assessment of their radiative transfer properties and also their potential to produce adverse health effects. In addition, the ability to predict or estimate the most relevant aggregate properties of these aerosols using approximations that agree well not only with experimental data but also with more detailed theoretical calculations is important in understanding the resulting impacts of these aerosols. In particular, The Rayleigh–Debye–Gans (RDG) approximation is routinely used to assess the angular distributions of scattered light and the mass scattering, mass extinction, and mass absorption coefficients for fractal aggregates [1–5].

The question naturally arises as to the accuracy of the RDG approximation and the ranges of validity relevant to size and

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morphology of the fractal aggregates. In order to address this question, experimental data were acquired for a range of fractal aggregate properties. Detailed numerical calculations of the scattering, extinction and absorption of a typical fractal aggregate were made as a function of size, morphology and chemistry (via the refractive index) using DDSCAT, an algorithm using the discrete dipole approximation (DDA) developed and maintained by Professor Bruce Draine at Princeton University [2]. The results of the DDA computations that best fit the experimental data were then compared to the RDG predictions using the same experimental data to assess the validity of this approximations and, if necessary, to adjust constant parameters within the RDG approximation to yield better agreement with the more detailed calculations and with the experimental data. This paper presents the most important results of this study.

2. Theory and experiment

The experiments are described in detail elsewhere [6,7], but briefly, combustible materials were allowed to burn or smolder in a small combustion chamber and the smoke that was generated flowed into a rectangular smoke chamber where two small mixing fans produced a homogeneous aerosol distribution. Samples were then flowed from the smoke chamber to various instruments for continuous recording of data. The data acquired included angular intensity data at four forward scattering angles (15°, 22½°, 30°, and 45°) and two backward scattering angles (135° and 150°) and at monochromatic wavelengths of 635 nm and 532 nm; total scattering efficiency at a wavelength of 520 nm and light extinction at a wavelength of 532 nm; total mass concentration of aerosol; and the output of a calibrated ionization chamber that allows for the determination of number mean diameter and aerosol number concentrations. In addition, filter samples were taken for correlation with the continuous aerosol mass measurements and additional filter samples obtained for subsequent analysis using a Scanning Electron Microscope (SEM) and Transmission Electron Microscope (TEM).

The angular intensity data were used to derive the radius of gyration, R_g, from the forward angle scattering measurements and the fractal dimension, D_f, from the 45° and two backward angle scattering measurements using the standard approximations. For the calibrated ionization chamber, the relevant equations are described elsewhere for the determination of number mean diameter, d_g, and number concentration of the aggregate aerosols, N [8]. Using the measured aerosol mass concentration, the average mass of an aggregate particle, M_a, can then be obtained from the simple relationship

$$M_a = 1 \times 10^{-9}\, M/N \tag{1}$$

where M is the mass concentration in mg/m^3 and N is the number concentration in particles/cm^3.

The mass of an aggregate containing n_p primary particles, each with diameter, d_p, and density, ρ_p, can also be expressed as

$$M_a = n_p \rho_p (\pi d_p^3/6) \tag{2}$$

For smoke (or soot) aggregates, the applicable fractal power law is

$$n_p = k_f (R_g/d_p)^{D_f} \tag{3}$$

where k_f is the prefactor with a value of approximately 6.45.

Substituting the expression for n_p (Eq. (3)) into Eq. (2), and rearranging, the following equation for d_p results

$$d_p = \left[(6/\pi)\left(M_a \Big/ \left(k_f \rho_p R_g^{D_f}\right)\right)\right]^{1/(3-D_f)} \tag{4}$$

Since all other variables appearing in Eq. (4) are known, then the primary particle diameter can be calculated in essentially real time. Determination of these parameter values is important for subsequent use in the DDA computations and in the RDG approximations.

For the RDG approximations, the following equations define the mass absorption, mass scattering and mass extinction coefficients.

$$\sigma_{abs} = 6\pi E_m/\lambda\rho_p \tag{5}$$

$$\sigma_{sca} = \left(4\pi n_2 (x_p)^3 F_m/\lambda\rho_p n_1\right)\left[1 + (4/3D_f)k^2 (R_g)^2\right]^{-D_f/2} \tag{6}$$

where E_m is the imaginary component of $(m^2-1)/(m^2+2) = Im[(m^2-1)/(m^2+2)]$, F_m is the square of the absolute value of $(m^2-1)/(m^2+2) = |(m^2-1)/(m^2+2)|^2$, m is the complex index of refraction, $m = n + ik_e$, x_p is the size parameter $= \pi d_p/\lambda$, where d_p is the diameter of a primary particle and k is the wave vector $= 2\pi/\lambda$, ρ_p is the density of a primary particle, taken to be 1.86 g/cm^3 and n_1 and n_2 are the first and second moments of the aggregate size distribution.

In the case of large aggregates, $n_2 \to (n_p)^2$ and $n_1 \to n_p$ and with $k^2(R_g)^2 \gg 1$, Eq. 16 reduces to

$$\sigma_{sca} = \left[4\pi (x_p)^3 k_f F_m/\lambda\rho_p\right]\left[3D_f/16(x_p)^2\right]^{D_f/2} \tag{7}$$

The mass extinction coefficient, σ_{ext}, is the sum of the mass absorption and mass scattering coefficients, or

$$\sigma_{ext} = \sigma_{sca} + \sigma_{abs} \tag{8}$$

The expressions used to estimate the RDG angular intensity distributions are described in detail in the literature and in the interest of brevity will not be presented here [9,10].

For the DDA calculations using the DDSCAT algorithm, a fractal aggregate consisting of 74 primary spherical particles was used. For input to DDSCAT, the coordinates of the center of each primary particle are specified, along with the radius of the primary particle, an index of refraction for the primary particles, the wavelength of incident electromagnetic radiation (EMR), and the volume equivalent radius, a_{eq}, of the aggregate. The algorithm then computes the intensity of scattered EMR for a multitude of different orientations of the aggregate with the incident beam and averages the scattered intensities over these multiple orientations to generate an average angular intensity distribution. DDSCAT also calculates the scattering, absorption, and extinction efficiencies for the aggregate for a given set of input conditions. For the base fractal aggregate, the primary particle radius is taken to be 0.50 units with the result that the primary particles barely touch each other at only a single point producing a chain-like aggregate with a fractal dimension of $D_f = 1.70$. By increasing the radius of each sphere while holding the coordinates of each sphere's center constant, the primary particles begin to overlap producing an aggregate that is more compact and less chain-like.

To determine the impact of the overlap on the morphology and volume equivalent radius of the aggregate, the coordinates of the 74 primary particles were entered into an AutoCad three dimensional space and the radius of each primary particle increased. AutoCad then produces the resultant image of the aggregate to show how its morphology has changed, but more than that, subroutines in AutoCad allow for the determination of the volume equivalent radius, surface equivalent radius and radius of gyration for the new overlapped particles that can be used as input into DDSCAT. To estimate the impact of overlap on the resultant fractal dimension, the data of Oh and Sorensen [11] was used that relates the increase in fractal dimension to the overlap, where the overlap, β, is defined as the ratio of an increased primary particle radius to

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