Supplementary Information

Investigation of Absorption and Scattering Properties of Soot Aggregates of Different Fractal Dimension at 532 nm Using RDG and GMM

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Besides the statistical scaling relationship, Eq. (1), it is also expected that the density autocorrelation function of finite-sized fractal aggregates have the following functional form (Filippov et al., 2000; Sorensen, 2001)

$$C(r) = Ar^{D_f - 3} h(r/\xi) \quad , \tag{S1}$$

where $C(r)$ is the density autocorrelation or two-point density-density correlation function, r is the distance variable, A is an appropriate constant, h is a cutoff function associated with the finite size of aggregates, and ξ is a characteristic length representing the size of the aggregate. The normalized density autocorrelation function can be viewed as the probability of finding a primary particle as a function of distance. Eq. (S1) implies that the autocorrelation function $C(r)$, when plotted on a log-log scale, should display a slope of (D_f-3) over an intermediate range of r for sufficiently large aggregates. For aggregates whose sizes are not large enough, the density autocorrelation function does not display the expected slope because of the influence of the cutoff function. It is noted that although the scaling law, Eq. (1), is satisfied intrinsically by all the numerically generated fractal aggregates using the tunable algorithm described earlier, there is no guarantee that the autocorrelation functions of these aggregates have the expected slope when plotted on a log-log scale over an intermediate distance. Therefore, it is important to check if the numerically generated aggregates have the expected property in terms of the slope of their autocorrelation functions. It is also well known that the autocorrelation function is directly linked to the light scattering behavior of fractal aggregates (Filippov et al., 2000; Sorensen, 2001).

The autocorrelation function was calculated as the distance distribution function described by Hasmy et al. (1993) and Filippov et al. (2000). Some typical results of normalized $C(r)$ vs. the non-dimensional distance r/d_p on log-log scale for N_p = 800 and different fractal dimensions are shown in Fig. S1. The three black straight lines have a slope of (D_f-3) with D_f being the respective value of the fractal dimension of the corresponding aggregate. Two observations can be made from Fig. S1. First, with increasing the fractal dimension the autocorrelation function first increases at smaller distances and then drops more rapidly at larger distances. This trend suggests that there exists more pairs of primary particle at a given small distance as the fractal dimension increases, which is expected since the structure of aggregate becomes more compact, i.e., the constituent primary particles are arranged closer to each other as D_f increases. Correspondingly, it is also expected that the autocorrelation function decreases more rapidly at larger distances as the aggregate becomes more compact, since the probability of finding pairs of primary particle at a given large distance diminishes. Secondly, the autocorrelation functions of the aggregates generated using the tunable cluster-cluster aggregation algorithm do display the expected slope on this plot over a range of intermediate distances, especially for D_f = 1.4 and 1.78. Although the slope of the autocorrelation function of D_f = 2.1 does not clearly display the expected slope, at least not over a well defined extended range of distance, it does not necessarily mean that the autocorrelation function of the aggregate for D_f = 2.1 and N_p = 800 does not have the expected slope. The reason for the not well established expected slope for the curve of D_f = 2.1 lies in the fact that the radius of gyration of this aggregate is not large enough and its density autocorrelation function is strongly affected by the cutoff function. This is why the expected slope of (D_f-3) for the curve of D_f = 2.1 in Fig. S1 is not yet well established. Based on the study of Filippov et al. (2000), who showed that the expected slope of (D_f-3) of the density autocorrelation function is gradually established as the aggregate size increases, it is expected that the correct slope of the density autocorrelation function of aggregates of D_f = 2.1 generated using the present tunable algorithm should be displayed for larger aggregates greater than N_p = 800. Nevertheless, it is reasonable to state that the results shown in Fig. S1 confirm that the numerically generated fractal aggregates using the present tunable algorithm indeed satisfy the fractal property given in Eq. (S1).

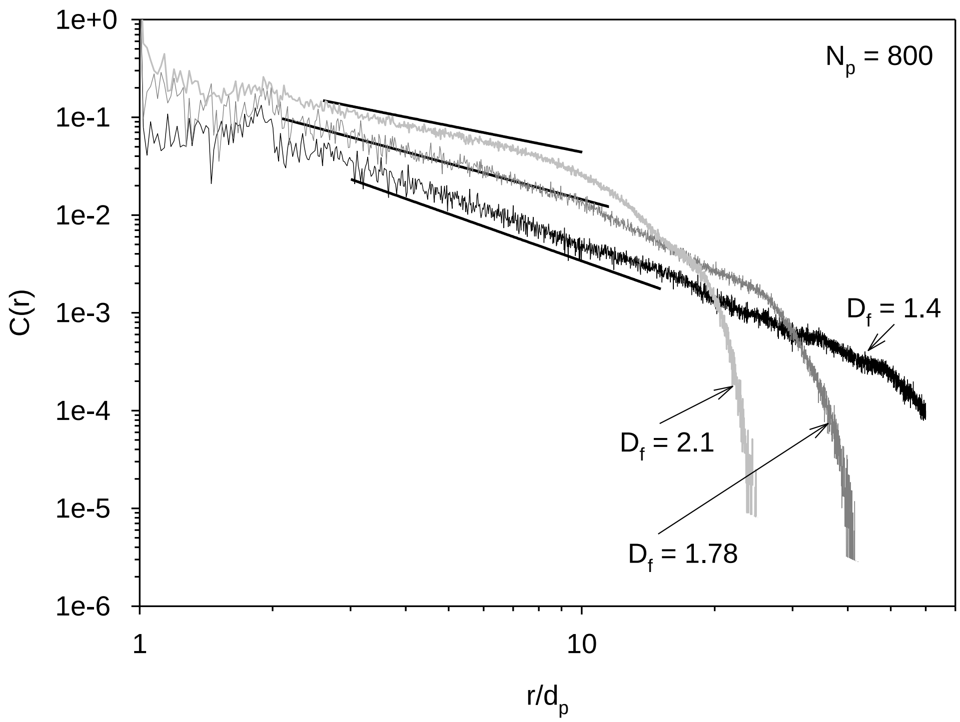

Fig. S1 Density autocorrelation functions of fractal aggregates generated using the following parameters: a = 15 nm k_f = 2.3, and three fractal dimensions of D_f = 1.4, 1.78, and 2.1 for aggregate size N_p = 800. The three thickened straight black lines have a slope of (D_f-3) on this log-log plot.