Supporting Online Material for

Fractal Organic Hazes Provided an Ultraviolet Shield for Early Earth

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Supporting Online Material

1 Materials and Methods

1.1 Model Basics

Simulations of the Archean haze were conducted using the Whole Atmosphere Community Climate Model (WACCM), a general circulation model, and the Community Aerosol and Radiation Model for Atmospheres (CARMA), a microphysical model. The model utilizes the sectional microphysical modeling capabilities of CARMA within the WACCM framework. WACCM version 3.1.9 was used along with CARMA version 2.3, which has been modified by the authors to simulate fractal aggregate particles. WACCM controls advection and wet deposition while CARMA controls production, vertical diffusion, coagulation, and dry deposition (S1). For the studies described in this work, the model is configured with 4°x5° horizontal resolution and 66 vertical levels extending to a maximum altitude of ~140 km. CARMA has been configured with 40 mass dependent bins, spanning aggregate radii from 1 nm to 1 mm. Haze aerosols are produced in a temporally and horizontally uniform manner and are initially sourced into the smallest mass bin (corresponding to $r = 1$ nm). The vertical distribution of haze production is specified by a user defined source function. Here, the haze source function is adapted from the photochemical modeling work of Pavlov et al. (S2). Pavlov et al. found that haze production occurred between altitudes of 40 and 80 km, peaking near 65 km. Here the haze source function is chosen to be lognormally distributed about a peak production altitude of 65 km (Fig. S1). Haze particles are assumed to have a material density of $\rho_m = 0.64$ g cm$^{-3}$ (S3). The thermal structure of the atmosphere is taken to be
that of the present day Earth and the haze particles are not allowed to radiatively
feedback on the model.

1.2 Fractal Microphysics

Fractal aggregates are composed of many primary spherical particles (called
monomers) that adhere together forming irregularly shaped, fluffy aggregates. In this
work the fractal dimension, $D_f$, and the monomer radius, $r_{mon}$, are free parameters. The
number of monomers per aggregate, $n_{mon}$, is proportional to the aggregate mass and thus
after appropriate selection of $D_f$ and $r_{mon}$, each aerosol mass bin can be assigned a
corresponding fractal radius, $R_f$. The fractal radius can be thought of as the geometric
collision radius of an aggregate particle. As such, $R_f$ is used in expressions for the
Knudsen number and for coagulation kernels. Note that the fractal radius of an aggregate
with $D_f < 3$ is always greater than the radius of an equal mass sphere, thus fractal
aggregates experience enhanced areas for coagulation and diminished non-continuum
effects when compared with spherical particles.

Use of the above defined fractal radius in Stokes law and in the calculation of
diffusion coefficients is not fully accurate. Fractal aggregates are permeable, thus fluid
can flow through their structure ($S4, S5$). This effect causes a decrease in the Stokes drag
force and an increase in the molecular diffusivity compared with predictions based on $R_f$.
Here, the aggregate permeability scheme suggested by Vainhstein et al. ($S6$) is used to
calculate the fractal mobility radius, $R_m$. The fractal mobility radius is the radius of an
equal mass sphere that experiences the same Stokes drag force as the aggregate in
question. The effect of permeability is most noticeable for smaller aggregates.
(n_{\text{mon}} < 500) with low fractal dimensions (D_f < 2) where R_m can be up to 25% less than R_f. The fractal mobility radius of an aggregate with D_f < 3 is always greater than the radius of an equal mass sphere, thus fractal aggregates experience reduced fall velocities and reduced diffusion coefficients when compared with spherical particles.

Studies of Titan hydrocarbon aerosols have assumed that all aggregates, regardless of size, have a fractal dimension of 2 (S7 - S11). This value is in general agreement with theoretical calculations of diffusion limited cluster-cluster aggregation in both the ballistic and continuum regimes. (S12, S13). Though valid as a first approximation, the assumption of a constant fractal dimension across all size bins misses an important nuance of fractal aggregate microphysics. Terrestrial carbonaceous fractal aggregate aerosols have been observed to restructure, becoming more compact (higher D_f) as the number of monomers contained in the aggregate increases (S14 - S17). The fractal dimension of early Earth (and Titan) haze aggregates therefore should not be considered constant, but rather will vary across the aggregate size distribution.

Hydrocarbon aerosols initially condense into nanometer sized spherical particles. The initial growth phase is characterized by a high fractal dimension (D_f \sim 3), as monomers are built molecule by molecule forming spherical particles with radii ranging from 10 to 100 nm. In the secondary growth phase, spherical monomers coagulate into short linear chains of low fractal dimension (n_{\text{mon}} < 100, D_f \sim 1.5). As aggregates grow large restructuring becomes important. Chain-like aggregates tend to be electrically charged. During the restructuring process oppositely charged aggregate limbs attach resulting in more compact arrangements. Laboratory experiments have shown that electrical restructuring increases the fractal dimension of carbonaceous aggregates to D_f \sim 2.4
Aggregate restructuring is also triggered by Brownian motion of monomers within an aggregate and by surface energy minimization of condensed water trapped within an aggregate (SI4, SI7). Restructuring is parameterized in the microphysical code by incorporating a size bin dependent fractal dimension (Fig. S2). The end result of the complex restructuring process is that the fractal dimension increases with aggregate mass.

2) Supporting Text

2.1 Haze Microphysical Properties

We performed a number of calculations to understand the sensitivity of the results reported in the main text to the underlying parameters. A summary of the basic parameters and pertinent results for each run is shown in table S1. Four runs were conducted using spherical microphysics. Six runs were conducted using fractal microphysics. Haze production rates, $\Phi$, were chosen to vary from $10^{12}$ to $10^{15}$ g yr$^{-1}$ in accordance with reasonable estimates for both the pre-biotic and post-biotic Earth (SI8). Simulations were run for six model years where upon the haze layer reaches steady-state. The data presented here are annual averages taken from the final year of the simulations.

In our simulations the highest particle number densities occur in the production region (Fig. S3). The haze particles grow rapidly due to coagulation in the production region where number densities are large and collisions frequent. Nanometer sized haze particles can diffuse upwards, while larger particles fall into the lower atmosphere where they continue to grow in size before eventually being removed by wet deposition in the troposphere or dry deposition at the ground. The annual mean zonally averaged haze mass concentration for the fractal model is plotted in figure S4. The haze is distributed in
a zonally uniform manner with higher mass concentrations over the poles than over equatorial regions. The steep horizontal gradient is caused by the Brewer-Dobson circulation which transports haze poleward where the haze particles then descend into the lower atmosphere. There is a slight bias towards mass loading over the southern hemisphere pole due to its stronger polar vortex that enhances transport (SI). We expect that the latitudinal gradients will be reduced in a simulation in which the haze is radiatively active (S19). The bulk of the haze mass is concentrated between 10 and 30 km altitude.

In this work, $r_{mon}$ is a free parameter that affects both the fractal microphysical and fractal optical models. A standard value of $r_{mon}$ is taken to be 50 nm (S1I), however values of 20 nm and 100 nm are also explored in limited cases to ensure the results presented here are not artifacts of convenient parameter selection. The haze effective optical depth as a function of wavelength is calculated using the model outputted particle size distributions in conjunction with the relevant optical model; Mie theory for the spherical model and the mean-field approximation for Mie scattering by aggregates of identical spheres for the fractal model (S20). In scattering atmospheres the extinction optical depth does not provide an accurate estimate of the attenuation of downwelling radiative flux. The extinction optical depth over estimates attenuation by neglecting forward scattered photons that remain in the downwelling beam. A more appropriate measure is the effective optical depth given by

$$\tau_{eff} = 1 - \frac{\omega}{2} - \frac{g\omega}{2} \tau_{ext}$$

(1)

where $g$ is the asymmetry parameter, $\omega$ is the single scattering albedo, and $\tau_{ext}$ is the extinction optical depth (S2I). We adopt the above formulation for the effective optical
depth given by Chylek and Wong rather than that of Rannou et al. (S8, S10) because the latter fails in the limit of a purely scattering atmosphere. The above formulation does not suffer such problems. Early Earth hazes are strongly forward scattering in the UV, thus $\tau_{\text{eff}}$ is only a few percent greater than the absorption optical depth, $\tau_{\text{abs}}$, in this regime. At infrared wavelengths, forward scattering becomes less pronounced and for Titan analog tholins $\omega$ trends toward 1. Then $\tau_{\text{eff}}$ can be an order of magnitude greater than $\tau_{\text{abs}}$. Visible wavelengths represent a transition region between the two regimes.

Fractal aggregate hazes have longer atmospheric lifetimes than do spherical particles. Aggregate sedimentation velocities are reduced as a result of their large mobility radii. Atmospheric lifetimes calculated for each simulation are listed in table S1. Haze particles have atmospheric lifetimes of up to ~3 years depending on the production rate. The atmospheric lifetime of haze particles decreases with increasing production rate in both fractal and spherical cases. Higher production rates lead to the formation of increasingly heavier haze particles that fall out of the atmosphere more rapidly regardless of the microphysical model used. In this study we test the haze production profile given by Pavlov et al. (S2), however changes in the production profile may alter our results. A haze production profile shifted to a greater altitude would likely increase atmospheric lifetime, particle size, and optical thickness. A haze production profile shifted to a lower altitude would likely reduce the aforementioned quantities. However, we expect such changes to be small since coagulation controls the haze microphysical properties in the lower stratosphere where the mass is greatest. The time for coagulation to occur is controlled by the time it takes haze particles to fall to the lower stratosphere, which is not strongly affected by the location of the production
region. The time to reach the lower stratosphere is nearly independent of the starting pressure because fall velocities are proportional to the inverse of the atmospheric density. We do not expect changes in the production profile to significantly affect the ability of a fractal haze to act as an ultraviolet shield as the unique radiative properties of a fractal haze are primarily attributed to the bimodal optical behavior of aggregates, which remains independent of reasonable changes in the haze production profile.

At each model grid point the average number of monomers per aggregate is calculated using the equation,

\[ n_{\text{mon}} = \frac{MD}{ND} \left( \frac{3}{4\pi \rho_m r_{\text{mon}}^3} \right) \]  \hfill (2)

where \( MD \) is the mass concentration, \( ND \) is the particle number density, \( \rho_m \) is the haze material density, and \( r_{\text{mon}} \) is the average monomer radii. \( MD \) and \( ND \) are outputted by the microphysical model. Fluffy micron sized aggregates containing large numbers of monomers dominate the lower atmosphere (Fig. S5). Above \( \sim 50 \text{ km} \) monomer and submonomer sized particles dominate the haze population. The number of monomers per aggregate increases with increasing haze production rate. In the lower atmosphere \( n_{\text{mon}} \sim 500 \) for \( \Phi = 10^{12} \text{ g yr}^{-1} \) while \( n_{\text{mon}} \sim 10^5 \) for \( \Phi = 10^{15} \text{ g yr}^{-1} \).

Varying the choice of \( r_{\text{mon}} \) has minimal effect on the total atmospheric mass, atmospheric lifetime, and the average mass per particle, however \( r_{\text{mon}} \) has a significant effect on the geometric size of aggregates. The similarity between the effective radii of equal mass spheres (Fig. S6a) for all choices of \( r_{\text{mon}} \) demonstrates that the average mass per particle is nearly identical in all cases. However, fractal effective radii vary significantly with \( r_{\text{mon}} \) (Fig. S6b). Since the average mass per particle is approximately
constant for all choices of $r_{\text{mon}}$, one can show that $R_f \propto r_{\text{mon}}^{(D_f-3)/D_f}$. For $D_f = 2$ this equation reduces to $R_f \propto r_{\text{mon}}^{1/2}$. In the lower atmosphere aggregates formed by 20 nm monomers have effective radii ~50% greater than aggregates formed by 50 nm monomers and double that of aggregates formed by 100 nm monomers. Figure S7 shows the average number of monomers per aggregate plotted as function of altitude for a haze production rate of $\Phi = 10^{14}$ g yr$^{-1}$ with average monomer radii of 20, 50 and 100 nm respectively. Since the average mass per aggregate is equal for all choices of $r_{\text{mon}}$, it follows that $n_{\text{mon}} \propto r_{\text{mon}}^{-3}$. The fractal aggregate optical model is dependent on both $r_{\text{mon}}$ and $n_{\text{mon}}$.

2.2 Haze Radiative Properties

The optical properties of fractal aggregates were determined using the mean-field approximation for Mie scattering by aggregates composed of identical spheres described by Botet et al. (S20). This method has been validated against exact solutions to Maxwell’s equations for scattering by aggregates composed of multiple spheres as described by Xu (S22). However, a validation was only conducted for a silica aggregate having $n_{\text{mon}} = 64$, a monomer optical size parameter of 3.9, and a complex refractive index of $1.4 + 0.0001i$. A rigorous validation for large absorbing aggregates typical of this study has not been conducted. It should be noted that Botet’s method follows directly from that of Xu, constructed with the same guiding principles but with an interest to increase computational speed. Xu’s solutions have been validated by laboratory experiments, but only for monomer optical size parameters of ~5, and monomer numbers of 2, 8, and 27 (S23). In our study, monomer optical size parameters at wavelengths of
interest are ~1 and monomer numbers reach ~10^4. Though a rigorous validation for large absorbing aggregates is lacking, it should be noted that the fractal optical model of Botet has had widespread success over the past decade in replicating the optical properties of Titan fractal hazes (S9, S10, S24). Early Earth fractal hazes were likely similar in both composition and structure to Titan fractal hazes. Here we have adopted optical constants for Titan analog haze particles (S25). The incorporation of oxygenated species into the haze has been shown to more than double the imaginary part of the refractive index (S26), however data is only available at a single wavelength (λ = 532 nm). It unclear how the wavelength dependence of optical constants for oxygenated haze particles would differ if at all from those derived for Titan analogs. We conducted a series of sensitivity studies that indicate the UV shielding characteristics of fractal aggregates are largely independent of the chosen refractive indices. Cases using imaginary refractive indices 5 times larger and 5 times smaller than those found by Khare et al. both indicate strong UV shielding by fractal aggregates (Fig. S8). Strong UV shielding was also found in a test case using a constant refractive index across all wavelengths (Fig. S9). The fractal optical model preserves UV shielding regardless of the wavelength dependence of the chosen optical constants. This is because the optical size parameter (the ratio of particle circumference to wavelength of light) for fractal aggregates is based on the monomer radius, not the total aggregate radius.

Figure S10 shows the asymmetry parameter for early Earth fractal haze particles plotted against wavelength. Fractal particles are slightly less forward scattering at short wavelengths while being more forward scattering at mid-visible wavelengths when compared to equal mass Mie particles. Figure S11 shows the single scattering albedo as a
function of wavelength for a 1000 monomer aggregate and an equal mass Mie particle. The aggregate particle is more absorbing than a Mie particle at wavelengths below 200 nm, while being more scattering in the mid-visible. The effect of fractal optics on the extinction and absorption coefficients is discussed in the main article.

As is evident in table S1, for all haze production rates a fractal aggregate haze is an order of magnitude or more optically thick in UV than in the visible when compared with a spherical haze. In both the spherical and fractal models, haze production rates of $10^{12}$ and $10^{13}$ g yr$^{-1}$ yield a steady state haze that is too optically thin to be of significant interest with respect to the faint young Sun paradox. In these cases both UV shielding and antigreenhouse cooling would be virtually non-existent and thus the haze would not perturb the Archean climate.

A haze production rate of $\Phi = 10^{15}$ g yr$^{-1}$ poses a potentially interesting though less likely view of the Archean climate. In this case both models yield a steady state haze that is thick in the visible with effective optical depths of 2.3 and 2.6 at $\lambda = 564$ nm for the spherical and fractal models respectively causing strong antigreenhouse cooling. However, a fractal haze would also be enormously thick in the UV ($\tau_{uv} \sim 70$) which would allow the build up of NH$_3$ and CH$_4$ to proceed virtually unhindered by photochemical destruction. If greenhouse gas concentrations were high enough, the young Earth could have warmed to above the freezing point despite strong antigreenhouse cooling. This scenario views the Archean atmosphere somewhat like that of Venus presently where intense greenhouse warming keeps the surface temperature high despite only a small fraction of the incident solar radiation actually reaching the surface (S27). However, in the absence of sufficient greenhouse gases, a thick haze may
have initiated the mid-Archean glacial interval circa 2.9 Ga as described in the work of Domagal-Goldman (S28).

A production rate of $10^{14}$ g yr$^{-1}$ provides the most interesting yet plausible scenario for the Archean climate. The results from these runs are the focus of the main article.

The radiative properties of the haze show some sensitivity to the free parameter $r_{mon}$, however the basic result holds true. Figure S12 shows the global mean effective optical depth versus wavelength for average monomer radii of 20, 50, and 100 nm. Both the 20 and 50 nm cases have similar wavelength dependence with $\tau_{uv}/\tau_{vis} \sim 22$. For the case of $r_{mon} = 100$ nm, the wavelength dependence of the optical depth is less drastic. However $r_{mon} = 100$ is more than double the proposed upper limit for average monomer radii proposed by Tomasko (S11) for Titan monomers and is not likely to represent reality. Though the magnitude of the optical depth is sensitive to the monomer radius, $\tau_{uv}/\tau_{vis}$ remains high in all cases, thus the choice of $r_{mon}$ does not affect the primary conclusions of this work. An additional simulation that assumed a constant fractal dimension of 2 did not yield meaningful differences from the results shown here either, indicating that our primary results are not affected by the reasonable choices of $D_f$.

In this study the haze layer does not radiatively feedback on the model. Heating of the haze layer would alter upper atmospheric dynamics. Fig. S13 shows the vertical profile of the globally averaged absorption coefficient in the UV and mid-visible. The peak of absorption occurs near an altitude of 20 km, which would cause a stratosphere to form even in the absence of ozone heating. Robock et al. (S29) showed that an absorption optical depth of less than 0.1 leads to heating of 50 K or more in the upper
atmosphere. Heating in the stratosphere would create rising motions that would increase the atmospheric lifetime of haze. The latitudinal variations in haze distribution would lead to enhanced heating over the summer pole that would in turn act to diminish equator to pole temperature gradients thus weakening upper atmospheric circulations. Therefore, dynamics would likely reduce the latitudinal gradient in the distribution of the haze mass. UV shielding and antigreenhouse cooling effects would likely be more globally uniform than indicated in this work (Fig. S14). However, if the haze does retain the steep latitudinal gradients shown in figure S14, the picture of the Archean climate is more complicated. Ultraviolet shielding, and antigreenhouse would both strongly depend on latitude, creating a complex climate system requiring a 3-D model to fully resolve.

Supporting References

S4. X.-Y. Li, B.E. Logan, Wat. Res. 35(14), 3373 (2001)
S24. A. Bellucci et al., Icarus 201, 198 (2009)
3) Supporting Figures and Captions

**Fig. S1:** The haze source function for the four production rates explored. The vertical profile of the haze source function has been adapted from the 1-D photochemical modeling work of Pavlov et al. (S2). In this study the haze is assumed be produced in a temporally and horizontally uniform manner.

**Fig. S2:** The fractal dimension of aggregate aerosols varies with the number of monomers per aggregate (or analogously with the aggregate mass). Initial growth is characterized by a high fractal dimension as nanometer sized particles coalesce to form spherical monomers. The second growth stage is characterized by a low fractal dimension as spherical monomers coagulate to form linear chain-like structures. As aggregates grow larger, they tend to collapse into more compact arrangements resulting in a higher fractal dimension. Illustrated is the parameterization used in this study.

**Fig. S3:** Zonal mean annual average number density for a haze production rate of $10^{14}$ g yr$^{-1}$ and $r_{mon} = 50$ nm. Particles are initially produced in the model with a radius of 1 nm. Haze number densities peak in the photochemical production zone where sub-monomer sized particles dominate the population. Outside the production zone number densities decrease rapidly due to coagulation of monomers to form larger particles.

**Fig. S4:** Zonal mean annual average haze mass concentration for haze production rate of $10^{14}$ g yr$^{-1}$ and $r_{mon} = 50$ nm. The haze mass distribution features steep latitudinal gradients with the bulk of the mass located over the polar regions. In our model the haze
layer remains thin over the equatorial regions, however we expect that radiative
feedbacks would smear out the distribution leading to a more latitudinally uniform
coverage of haze.

**Fig. S5:** The average number of monomers per aggregate plotted versus height for haze
production rates ranging from $10^{12}$ to $10^{15}$ g yr$^{-1}$ and $r_{\text{mon}} = 50$ nm. Larger production
rates lead to higher monomer numbers.

**Fig. S6:** (A) The equal mass spherical effective radii calculated for aggregate particles for
a haze production rate of $10^{14}$ g yr$^{-1}$ and $r_{\text{mon}} = 20$, 50, and 100 nm respectively. The
average mass per particle remains relatively unchanged for all choices of $r_{\text{mon}}$. (B) The
fractal effective radius is sensitive to the choice of $r_{\text{mon}}$. A smaller choice for the average
monomer size yields larger aggregates for a given haze production rate.

**Fig. S7:** The number of monomers per aggregate for a haze production rate of $10^{14}$ g yr$^{-1}$
and $r_{\text{mon}} = 20$, 50, and 100 nm. A smaller choice for $r_{\text{mon}}$ yields a larger number of
monomers per aggregate.

**Fig. S8:** The ratio of absorption efficiencies calculated for fractal aggregate particles to
those calculated for equal mass Mie particles. A value of unity indicates that the
aggregate particle is radiatively indistinguishable from the Mie particle. The particles
illustrated have $D_f = 2$, $r_{\text{mon}} = 50$ nm, and $n_{\text{mon}} = 1000$. Cases are shown corresponding to
refractive indices for Titan analogue hazes determined by Khare *et al.* (S25), five times
greater Khare’s values, and five times smaller. In each instance aggregates absorb strongly in the UV while remaining relatively transparent in the mid-visible.

**Fig. S9:** The ratio of absorption efficiencies calculated for fractal aggregate particles to those calculated for equal mass Mie particles. A value of unity indicates that the aggregate particle is radiatively indistinguishable from the Mie particle. The particles illustrated have $D_f = 2$, $r_{mon} = 50$ nm, and $n_{mon} = 10, 100, \text{and 1000}$ corresponding to fractal radii of 0.16 µm, 0.5 µm and 1.6 µm respectively. A constant refractive index of $n = 1.70 + i0.023$ is assumed at all wavelengths. Even after removing the wavelength dependence of refractive indices, fractal aggregate particles are more absorbing in the UV while being slightly less absorbing in the visible than equal mass Mie particles. This is due to the strong interaction of monomers contained in the aggregate with shortwave radiation.

**Fig. S10:** The asymmetry parameter for a fractal aggregate with, $n_{mon} = 1000$, $r_{mon} = 50$ nm, and $D_f = 2$ and for an equal mass Mie particle plotted against wavelength of incident radiation.

**Fig. S11:** The single scattering albedo for a fractal aggregate with $n_{mon} = 1000$, $r_{mon} = 50$, and $D_f = 2$ and for an equal mass Mie particle plotted against wavelength of incident radiation.
**Fig. S12:** The effective optical depth ($\tau_{eff}$) plotted for the fractal models for $\Phi = 10^{14} \text{ g yr}^{-1}$ and $r_{mon} = 20, 50, \text{ and } 100 \text{ nm}$ respectively. Though subtle variations are present, in all cases the haze layer is thick in the UV while remaining relatively thin in the visible. The choice of $r_{mon}$ is not found to affect the primary results of this work.

**Fig. S13:** The global mean vertical profile of the absorption coefficient is plotted versus altitude for a haze production rate of $10^{14} \text{ g yr}^{-1}$ and $r_{mon} = 50 \text{ nm}$ using the fractal model. Absorption is strongest near 20 km. The haze layer would strongly heat the air and create a stratosphere.

**Fig. S14:** The latitudinal distribution of the effective optical depth. In our model the haze layer is an order of magnitude thicker over the poles than over the equator. It is likely that radiative feedbacks of the haze on dynamics, not included in our model, would significantly reduce latitudinal gradients creating a more globally uniform haze layer.
**Table S1**: A summary of the model runs conducted in this work. For all production rates a fractal aggregate haze layer is an order of magnitude or more thicker in the UV than in the visible.

<table>
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<th>production rate $\Phi$ (g yr$^{-1}$)</th>
<th>model</th>
<th>$r_{\text{mon}}$ (nm)</th>
<th>atmospheric lifetime (yrs)</th>
<th>$\tau_{\text{uv}}$ (197 nm)</th>
<th>$\tau_{\text{vis}}$ (564 nm)</th>
<th>$\tau_{\text{uv}} / \tau_{\text{vis}}$</th>
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Fig. S2

The graph illustrates the relationship between the fractal dimension, $D_f$, and the number of monomers, $n_{mon}$, with three distinct phases:

1. **Monomer Growth**: The fractal dimension starts at a high value and decreases sharply as the number of monomers increases, indicating a transition from a compact to a more expanded structure.

2. **Aggregate Restructuring**: This phase shows a dramatic change in the fractal dimension, indicating a restructuring or reorganization of the aggregate structure.

3. **Chain Growth**: As the number of monomers increases further, the fractal dimension levels off, suggesting a more established chain-like structure.

The x-axis represents the number of monomers, while the y-axis represents the fractal dimension. The graph highlights the dynamic changes in structure as the system evolves from monomer growth to aggregate restructuring and finally to chain growth.
Fig. S4
Fig. S5
Fig. S6
Fig. S7
Fig. S9

- $D_t = 2$
- $r_{\text{mon}} = 50 \text{ nm}$
- $n = 1.70 \pm 0.023$

The graph shows the change in $Q_{\text{fracal}}/Q_{\text{in}}$ with wavelength ($\mu$m) for different values of $r_{\text{mon}}$: 1000, 100, and 10.
Fig. S10

The diagram shows the asymmetry parameter (g) as a function of wavelength (μm). Two lines are plotted:

- Red line: fractal, $n_{\text{mon}} = 1000$
- Blue line: equal mass Mie

The asymmetry parameter decreases with increasing wavelength, with the fractal line starting higher than the equal mass Mie line and the equal mass Mie line ending lower than the fractal line.
Fig S11
Fig. S12

The figure shows the effective optical depth, $\tau_{ef}$, as a function of wavelength ($\mu$m) for different monomer radii, $r_{mon}$. The curves represent $r_{mon} = 20$, $50$, and $100$, respectively.
Fig. S14

![Graph showing effective optical depth, $\tau_{\text{opt}}$, vs. latitude (deg) for different particle properties. The graph includes curves for 197 nm, spherical; 584 nm, spherical; 197 nm, fractal, $r_{\text{min}} = 50$ nm; and 584 nm, fractal, $r_{\text{min}} = 50$ nm.](image-url)