

Response for Review of: “Fractal Geometry of Aerosol Particles and Its Impact on Atmospheric Optical Properties: Development and Validation of the Fractal Aerosol Cluster Model in a Heavy Haze Event” by Zhexin et al.

The paper discusses a modeling effort to account better for the fractal nature of some atmospheric particles by coupling a fractal formulation called FACM (perhaps an unfortunate acronym choice) with WRF-Chem. The study results seem to suggest a significant improvement in visibility and surface radiation estimates when compared to surface measurements, at least in areas affected by combustion pollution and dry climate. I think the results are interesting and the work is worth publication after a few changes are made.

General comments:

1. The FACM model is not a new development on its own; it is based on established physics. What I think should be underlined, in terms of novelty, is more the coupling of WRF-Chem with FACM than FACM itself.

Reply to General Comment 1

We fully agree with the reviewer that the physical framework underlying FACM is not entirely new; the fractal structure of aerosol aggregates has been extensively studied through both observations and theoretical work. The novelty of this study lies primarily in translating this physical framework into an online-coupled parameterization scheme and, for the first time, implementing its coupling with WRF-Chem and validating it against observations. We have made this point more explicit in both the Introduction and the Conclusions.

Revision:

- (1) In the Introduction, we add the following sentences:

In summary, this study advances beyond prior work on fractal aggregates in several key aspects. **While the physical basis of fractal aerosol clusters is well established from field and laboratory studies, the novelty of this work lies in translating this physical framework into an online-coupled parameterization scheme.** First, we develop the Fractal Aerosol Cluster Model (FACM),...

- (2) In the Conclusions, we add the following sentences:

... , with station observations used for validation. **While the physical basis of fractal aerosol aggregates has been established through extensive field and laboratory studies, the novelty of this work lies in translating this physical framework into an online-coupled parameterization scheme and, for the first time, coupling it with WRF-Chem for validation against observations.** The results for atmospheric visibility and surface shortwave radiation flux demonstrated the superior performance of EXP compared to CTR. The main findings of this study are as follows:

2. A lot of acronyms used in the abstract make it a bit difficult to read.

Reply to General Comment 2

We thank the reviewer for this helpful comment. We agree that the abstract contained too many acronyms, which made it less readable. In the revised abstract, we have minimized the use of acronyms: AV and OBS have been replaced by “atmospheric visibility” and “observations,” respectively; EXP and CTR have been replaced by descriptive terms (“the experimental simulation” and “the control simulation”). Only widely recognized or essential abbreviations (FACM, WRF-Chem, and PM_{2.5}) are retained, with their full names given at first appearance. The revised abstract is provided below.

Revision:

Replace the entire abstract with the following revised version:

Aerosol particles, as crucial atmospheric components, significantly influence optical properties and play an important role in climate change. Based on Lorenz-Mie theory, the optical equivalent radius of aerosol particles affects atmospheric visibility. Traditional aerosol models, which assume spherical or other geometrically regular particle shapes, significantly deviate from observations in visibility simulations. In this study, the mechanism of random diffusion and aggregation of monomeric particles into fractal geometrical clusters was reproduced. The Fractal Aerosol Cluster Model (FACM) was developed to parameterize the optical and aerodynamic sizes of the fractal geometry of aerosol particles. Sensitivity experiments were conducted to simulate a severe haze event in northern China in November 2018 by coupling FACM into WRF-Chem. The simulated near-ground PM_{2.5} mass concentrations in the experimental and control simulations are similar to the observations. However, the experimental simulation produced larger extinction coefficients and lower atmospheric visibility, which is closer to the observations. The average normalized mean error of atmospheric visibility in the experimental simulation is 163%, compared to 422% in the control simulation. Thus, considering the fractal geometry of aerosol particles significantly improves simulated atmospheric visibility. Furthermore, a reduction of approximately 60 W m⁻² in land surface shortwave radiation in the experimental simulation compared to the control simulation was also confirmed by observations. This study of the optical properties of fractal aerosol clusters will contribute to future research on atmospheric environment and climate forcing.

3. The literature cited seems quite limited and often narrowly chosen, with perhaps some biases.

Reply to General Comment 3

We thank the reviewer for this comment and agree that the original citation list could be broadened. In the revised manuscript, we have expanded the references substantially in response to this general comment and to the specific comments below. Both seminal early works and recent studies have been added in the Introduction, Section 2.1, Section

2.2, Section 3, and Section 4. The specific additions are detailed in our point-by-point responses to the relevant comments (e.g., Lines 89–96, 97–99, 171–173, 184, 447–450, and 628–630) and are reflected in the updated reference list. These additions broaden the literature coverage without altering the narrative logic of the paper.

In addition, Supplementary Material S2 presents preliminary sensitivity experiments over East Asia, which further support the transferability of FACM and are referenced in the revised Conclusions.

Revision:

References have been added at the specific locations indicated in the point-by-point responses below. No structural changes to the manuscript were required. The full set of newly added references is provided in the revised reference list.

- (3) The paper is overall clearly written except for a few grammar issues, but it could be more succinct; in some sections, there are a few repetitions that could be avoided.

Reply to General Comment 4

We thank the reviewer for this helpful comment. We have carefully proofread the entire manuscript for grammar and language issues and have removed redundant expressions to improve conciseness. The revised manuscript has been polished throughout. Specific changes are reflected in the revised manuscript.

Revision:

The entire manuscript has been language-polished and redundancies removed. Please refer to the revised manuscript for details.

4. The description of the choice of $D_f=2.7$ should instead be more extensive because that's key to the findings discussed here. More on this issue below.

Reply to General Comment 5

We thank the reviewer for emphasizing this point. We agree that the choice of $D_f = 2.7$ is central to interpreting the results presented in this study. In the revised manuscript, we have substantially expanded the description of the basis for selecting $D_f = 2.7$. The revised text now explains that this value should be understood as an effective fractal dimension representing the overall aerosol population, rather than a value derived from a specific aerosol composition ratio. It also clarifies that the real atmospheric average D_f is expected to lie between 2.1 (freshly formed, freely coagulated soot aggregates) and 3.0 (fully compact spheres), with aging processes driving D_f toward 3.

The choice of 2.7 was determined through sensitivity experiments comparing $D_f = 3.0, 2.7, 2.4$, and 2.1 which are presented in Supplementary Material S1. Those experiments show that $D_f = 2.7$ gives results closest to observations, whereas lower values (2.4 and 2.1) lead to AOD and VIS changes that far exceed the range supported by observations. Thus, $D_f = 2.7$ is a reasonable and conservative value based on

observational constraints and repeated tests, rather than a precise derivation.

Revision:

The paragraph describing the choice of $D_f = 2.7$ (previously at lines 437–438) has been replaced by an expanded version, as detailed in our response to the specific comment on lines 437–438. That expanded version now appears in the revised manuscript (Section 2.1). In addition, a cross-reference to Supplementary Material S1 has been added where the sensitivity experiments are discussed.

Specific comments:

1. Abstract, line 39: “more closely” -> “closer”

Reply to Specific Comment 1

We thank the reviewer for this correction. The phrase “more closely” has been changed to “**closer**” in the revised abstract.

2. Abstract, line 40: Can the authors really claim to know the differences with all these digits: 163.39% and 421.62%? Maybe 163% and 422% would be more than enough.

摘要第 40 行：作者真的能声称知道这些数字的差异：163.39%和 421.62%吗？也许 163%和 422%就足够了。

Reply to Specific Comment 2:

We agree with the reviewer that reporting the normalized mean error with two decimal places is unnecessary. These values were originally retained with two decimals without considering the appropriate precision for this metric. We have rounded them to 163% and 422%, respectively.

Revision:

In the abstract, “**The average normalized mean error of atmospheric visibility by EXP is 163.39%, compared to 421.62% by CTR**” has been replaced by “**The average normalized mean error of atmospheric visibility in the experimental simulation is 163%, compared to 422% in the control simulation.**”

3. Lines 78-81: The sentence is somewhat unclear. Also, remove the space at the end.

Reply to Specific Comment 3:

We thank the reviewer for pointing this out. The original sentence used the abbreviation “AV” without prior definition in the abstract, which made it unclear. We have replaced “AV” with “atmospheric visibility (AV)” at its first occurrence and removed the trailing space at the end of the sentence.

Revision:

The sentence “**simulated AV often exhibits large discrepancies from observations, with errors reaching up to 60%**”

has been replaced by “**simulated atmospheric visibility (AV) often exhibits large discrepancies from observations, with errors reaching up to 60%**”

4. Lines 89 to 96: The limited references cited here barely scratch the surface of the vast literature in this field and leave out some seminal papers on the topic.

Reply to the comment 4:

We thank the reviewer for pointing out the limited citation coverage in this paragraph. We agree that the original list did not adequately reflect the extensive literature on this topic. In the revised manuscript, we have added several seminal references to this section, covering early studies on the fractal nature of smoke aggregates, the development of the diffusion-limited aggregation model, foundational reviews of light scattering by fractal aggregates, and key works on the optical modeling of black carbon. The newly added references in this paragraph are:

- Forrest, S. R., & Witten, T. A. (1979). Long-range correlations in smoke-particle aggregates. *Journal of Physics A: Mathematical and General*, 12(5), L109–L117.
- Meakin, P. (1983). Formation of fractal clusters and networks by irreversible diffusion-limited aggregation. *Physical Review Letters*, 51(13), 1119–1122.
- Mackowski, D. W., & Mishchenko, M. I. (1996). Calculation of the T matrix and the scattering matrix for ensembles of spheres. *Journal of the Optical Society of America A*, 13(11), 2266–2278.
- Sorensen, C. M. (2001). Light scattering by fractal aggregates: A review. *Aerosol Science and Technology*, 35(2), 648–687.
- Kahnert, M., & Kangers, F. (2020). Modelling optical properties of atmospheric black carbon aerosols. *Journal of Quantitative Spectroscopy and Radiative Transfer*, 244, 106849.

These citations have been inserted at the appropriate places in the paragraph discussing non-spherical representations and fractal geometry.

5. Lines 97-99: I would say this statement is incorrect. Many, many papers discuss the detailed treatment of fractal aggregates with various degrees of approximation, up to very high.

Reply to the comment 5:

We thank the reviewer for this careful reading. We agree that the original statement was too sweeping: many studies have indeed treated fractal aggregates with varying degrees of approximation, including highly detailed optical calculations. In the revised manuscript, we have reformulated this passage to acknowledge these advances while clarifying the remaining gap specifically for online-coupled regional models.

The revised text now is as following:

Despite these advances, a gap remains for online-coupled regional models: detailed optical calculations for fractal aggregates (e.g., DDA, T-matrix) are computationally expensive and typically applied offline to individual aggregates (Mishchenko et al.,

1996; Mackowski & Mishchenko, 1996; Liu et al., 2018; Kahnert & Kanngießer, 2020), while most existing parameterizations lack a direct, analytically tractable link between the fractal growth process and the resulting optical and aerodynamic properties. Moreover, most fractal aerosol schemes have not been online coupled into regional meteorology–chemistry models, limiting their ability to capture the fully interactive aerosol–radiation–meteorology feedbacks that are crucial during heavy haze events.

Newly added references in this passage:

- Mishchenko, M. I., Travis, L. D., & Mackowski, D. W. (1996). T-matrix computations of light scattering by nonspherical particles: A review. *Journal of Quantitative Spectroscopy and Radiative Transfer*, 55(5), 535–575.
- Mackowski, D. W., & Mishchenko, M. I. (1996). Calculation of the T matrix and the scattering matrix for ensembles of spheres. *Journal of the Optical Society of America A*, 13(11), 2266–2278.
- Liu, C., Teng, S., Zhu, Y., et al. (2018). Performance of the discrete dipole approximation for optical properties of black carbon aggregates. *Journal of Quantitative Spectroscopy and Radiative Transfer*, 221, 98–109.
- Kahnert, M., & Kanngießer, F. (2020). Modelling optical properties of atmospheric black carbon aerosols. *Journal of Quantitative Spectroscopy and Radiative Transfer*, 244, 106849.

These citations support the statement that detailed optical methods exist but are computationally expensive and typically applied offline.

6. Lines 171-173: There are several other laboratory studies showing this process.

Reply to the comment 6:

We thank the reviewer for pointing this out. We agree that additional laboratory studies have documented the compaction of fractal aggregates during aging. In the revised manuscript, we have added several references to support this statement.

Newly added references:

- Mikhailov, E. F., Vlasenko, S. S., Podgorny, I. A., et al. (2006). Optical properties of soot-water drop agglomerates: An experimental study. *Journal of Geophysical Research: Atmospheres*, 111(D7), D07209. doi:10.1029/2005JD006389
- Zhang, R., Khalizov, A. F., Pagels, J., et al. (2008). Variability in morphology, hygroscopicity, and optical properties of soot aerosols during atmospheric processing. *Proceedings of the National Academy of Sciences*, 105(30), 10291-10296. doi:10.1073/pnas.0804860105
- Xue, H., Khalizov, A., Wang, L., et al. (2009). Effects of Dicarboxylic Acid Coating on the Optical Properties of Soot. *Physical chemistry chemical physics : PCCP*, 11, 7869-7875. doi:10.1039/b904129
- Zelenay, V., Mooser, R., Tritscher, T., et al. (2011). Aging induced changes on NEXAFS fingerprints in individual combustion particles. *Atmos. Chem. Phys.*, 11(22), 11777-11791. doi:10.5194/acp-11-11777-2011

7. Line 184: Provide citations for these observations.

Reply to the comment 7:

We thank the reviewer for this suggestion. We have added appropriate citations for the box-counting method used to calculate the fractal dimension.

Revision:

The figure caption:

d: the fractal dimension of non-spherical fractal cluster above by Box-counting method, given by the slope of the linear fitting, side length of each three-dimensional box was denoted by l .

has been revised to:

d: the fractal dimension of the non-spherical fractal cluster obtained by the box-counting method (Liebovitch & Toth, 1989; Forrest & Witten, 1979), given by the slope of the linear fitting; the side length of each three-dimensional box is denoted by l .

Newly added references:

Liebovitch, L. S., & Toth, T. (1989). A fast algorithm to determine fractal dimensions by box counting. *Physics Letters A*, 141(8–9), 386–390.

Forrest, S. R., & Witten, T. A. (1979). Long-range correlations in smoke-particle aggregates. *Journal of Physics A: Mathematical and General*, 12(5), L109–L117.

8. Line 200: Where does the sqrt of 3/5 come from?

Reply to the comment 8:

We thank the reviewer for this question. We would like to clarify that the factor $\sqrt{3/5}$ does not originate from fractal scaling theory. It comes from the classical result for the radius of gyration of a uniform solid sphere in mechanics (Rubinstein & Colby, 2003). For a uniform solid sphere of radius R , the radius of gyration R_g is defined as:

$$R_g^2 = \frac{\int r^2 dV}{\int dV} = \frac{\int_0^R r^2 \cdot 4\pi r^2 dr}{\frac{4}{3}\pi R^3} = \frac{3}{5} R^2$$

which gives

$$R_g = \sqrt{\frac{3}{5}} R,$$

For a uniform sphere, the geometric radius R is identical to the volume-equivalent radius R_{ve} , so the relation can also be written as

$$R_g = \sqrt{\frac{3}{5}} R_{ve}$$

This is a standard result in polymer physics, small-angle scattering (SAXS/SANS), and colloid science (Rubinstein & Colby, 2003). It is important to emphasize that this relation applies only to a continuous, uniform solid sphere, and not to fractal aggregates.

For fractal clusters, the relationship between R_g and R_{ve} is governed by the fractal scaling law, rather than by this constant factor. We mention $\sqrt{3/5}$ in the main text only to illustrate that constant factors appearing in the sphere limit ($D_f = 3$) do not affect the scaling exponent.

We have added a clarifying phrase in the main text to indicate the origin of this factor.

“...constant factors (such as k_0 or $\sqrt{3/5}$, the latter originating from the classical result for a uniform solid sphere, $R_g = \sqrt{3/5} R_{ve}$) do not affect the scaling exponent.”

9. Line 203: Does $D_f=3$ have to correspond to a sphere or just to a particle without random voids? For example, what is the D_f of a cube or an ellipsoid?

Reply to the comments 9:

We thank the reviewer for these two insightful questions.

$D_f = 3$ does **not** necessarily correspond to a sphere. The fractal dimension describes how the mass (or volume) of an object scales with its characteristic size. For any homogeneous, compact three-dimensional object without internal voids—such as a sphere, a cube, or an ellipsoid—the mass scales with the cube of the characteristic size, and therefore $D_f = 3$. In other words, $D_f = 3$ represents the class of **compact three-dimensional objects with no random internal voids**, rather than a specific shape. The spherical assumption is simply the most convenient geometric idealization used in traditional optical models, but it is not a requirement for $D_f = 3$.

10. Line 205: Similarly, does a $D_f < 3$ mean that the object is necessarily fractal?

Reply to the comments 10:

In fractal geometry, the fractal dimension is defined as (BATTY & LONGLEY, 1994; Mandelbrot, 1983)

$$D_f = \lim_{\varepsilon \rightarrow 0} \frac{\ln N(\varepsilon)}{-\ln \varepsilon},$$

where $N(\varepsilon)$ is the number of boxes of side length ε needed to cover the object. This definition characterizes how the measure of the object grows with scale, rather than its specific shape. When $D_f < 3$, the mass of the object grows more slowly with its characteristic size than for a compact three-dimensional body, which typically arises from the presence of internal voids or self-similar structures. In the context of

atmospheric aerosols, $D_f < 3$ is widely used as an indicator of fractal-like aggregates, such as soot clusters.

However, from a strictly mathematical perspective, $D_f < 3$ is not a sufficient condition for an object to be strictly self-similar or fractal in the classical sense. Conversely, one can construct mathematical objects that have an integer fractal dimension (e.g., $D_f = 1$ or 2) yet exhibit a fractal-like appearance. In the real atmosphere, whether such objects exist is still an open question. In this study, we use $D_f < 3$ as a practical and physically meaningful indicator of fractal morphology for aerosol aggregates, consistent with the observational and modeling literature.

So, in Section 2.2, immediately after the sentence introducing the fractal dimension and the spherical limit ($D_f = 3$), we have added a clarifying sentence:

“It should be noted that $D_f = 3$ does not correspond exclusively to a sphere; it represents any compact three-dimensional object without random internal voids, such as a cube or an ellipsoid. Conversely, $D_f < 3$ is used here as an indicator of fractal-like aggregates, although strictly speaking it is not a sufficient condition for fractal self-similarity in the mathematical sense.”

11. Line 229: A more careful definition of V_{tot} would be useful. How is it defined? By the smallest convex solid that fully encloses the particle? Or something different? From equation 4, it seems like that's the volume equivalent of a solid with the same radius of gyration.

Reply to the comments 11: definition of V_{tot}

We thank the reviewer for pointing out that a more careful definition of V_{tot} would be useful. We clarify its physical meaning, definition, and relation to the radius of gyration below.

(1) Physical definition of V_{tot} : In this study, V_{tot} is defined as the **volume enclosed by the outer envelope of a fractal aerosol cluster**, including both the material volume of all primary particles (V_m) and the void volume between them (V_{void}). Thus, $V_{\text{tot}} = V_m + V_{\text{void}}$, as shown in Eq. (3).

(2) On the “smallest convex solid” interpretation: We note the reviewer's suggestion of “the smallest convex solid that fully encloses the particle.” From the perspective of fractal geometry, the measure of a fractal object depends on the measurement scale. Therefore, it is difficult to define a strict minimum; the outer envelope is not necessarily convex, and it is not merely a solid. A more precise physical description is: **the total volume of the three-dimensional space enclosed by the outer envelope of an aerosol cluster when the envelope is described at a resolution res ($\text{res} \gg a$, where a is the**

equivalent radius of the primary particles comprising the cluster). This is the interpretation adopted in this study.

(3) Equation (4), $V_{\text{tot}} = \frac{4}{3}\pi R_g^3$, is an approximate expression for the envelope volume defined above. We consider this expression to be consistent, at least in order of magnitude, with “the volume equivalent to that of a solid with a radius equal to the radius of gyration.” It should be noted that we currently assume **isotropic growth** of the aerosol cluster, so that its outer envelope can be approximated as a sphere (similar to a fluffy spherical cotton ball). This assumption allows V_{tot} to be approximated using R_g as the characteristic scale.

(4) In reality, cluster growth is not necessarily isotropic; numerous observational studies have revealed diverse morphologies of aerosol aggregates. Therefore, approximating the outer envelope as a sphere may be a somewhat strong assumption. This point has not been further explored in the present study, but it is acknowledged in the outlook, and we plan to consider the effects of anisotropic growth on the envelope volume and fractal characteristics in future work.

Revision: have revised the explanatory text regarding eq. 3 as follows:

Here, $V_{\text{tot}} = V_{\text{void}} + V_m$ denotes the total volume of an aerosol cluster, defined as the volume enclosed by the outer envelope of the cluster at a resolution res ($\text{res} \gg a$, where a is the equivalent radius of the primary particles), which includes both the material volume V_m and the void volume V_{void} . Assuming isotropic growth, this envelope can be approximated as a sphere, and V_{tot} can be expressed in terms of the gyration radius as:

$$V_{\text{tot}} = \frac{4}{3}\pi R_g^3 \quad \text{eq. 4}$$

It should be noted that this spherical approximation relies on the assumption of isotropic cluster growth, whereas in reality aerosol aggregates may exhibit more diverse morphologies. This limitation is acknowledged in the outlook of this study.

12. Line 264 (and corresponding symbol in equation 8): how are vTs (mentioned in line 257, and then not used) and v different from each other?

Reply to the comments 12:

We thank the reviewer for catching this. It was a typographical error: Eq. (8) used v instead of v_{TS} , and the symbol definition also mistakenly defined v as the terminal settling velocity. We have corrected Eq. (8) and the symbol definition to consistently use v_{TS} throughout the manuscript.

Revision:

Revise v to v_{TS} in eq. 8 and the symbol definition after Eq. (8). Then full manuscript checked to ensure consistent use of v_{TS} .

13. Note, line 319: This seems like a strange approximation; what is the benefit of approximating 1.4 with 1? Also, a 40% difference does not seem negligible; certainly, it would not be for optical calculations using Mie, for example. So, I don't understand the justification or the advantage of such an approximation.

Reply to the comments 13:

We thank the reviewer for this question. We clarify the justification and advantages of this simplification below.

(1) The origin of the 1.4 coefficient is from (Rogak, Flagan, & Nguyen, 1993), in this paper, they derived an approximate relationship between the projected area equivalent diameter and the radius of gyration through numerical simulations and experimental measurements. In their model framework, this relationship is expressed as:

$$d_p \approx 1.4R_g \text{ for } D_f \gtrsim 2$$

The core conclusion of that study is that for aggregates with fractal dimensions close to or greater than 2, an approximately linear relationship exists between the projected area equivalent diameter and the radius of gyration, and the proportionality coefficient exhibits weak dependence on cluster size and monomer number.

(2) Simplifying to 1 does not change the core scaling conclusions of this study. The central goal of this study is to establish power-law relationships between D_f and the optical/aerodynamic equivalent radii. In scaling analysis, any constant factor independent of N and D_f is absorbed into the prefactor and does not affect the power-law exponent. Whether the prefactor is 1.4 or 1, the scaling behaviors $R_{oe} \propto N^{1/D_f}$ and $R_{ae} \propto N^{(D_f-1)/(2D_f)}$ remain completely unchanged.

Notably, in fractal aerosol optical parameterization studies, the optical properties are known to be much less sensitive to the prefactor (K_f) than to the fractal dimension (D_f). For example, (J. Liu, Zhu, Zhou, & Han, 2024) explicitly state that the absorption properties are much less sensitive to K_f than to D_f , and therefore fix K_f at a constant value of 1.20 (rather than 1.4), while allowing D_f to vary over the range 1.80–2.80. (Romshoo et al., 2021) even omit discussion of K_f entirely. Liu et al. (F.-S. Liu, Snelling, & Smallwood, 2009) also specifically investigated the effect of the prefactor on the optical properties of fractal soot aggregates and found its influence to be limited. Therefore, simplifying 1.4 to 1 is consistent with common practice in this field.

(3) The projected area equivalent diameter formula proposed in (Rogak et al., 1993) is a piecewise parameterized model covering structures ranging from linear chains to

compact spheres, with the parameter $\alpha_p = 0.67$ used to describe the linear chain limit (<https://utoronto.scholaris.ca/server/api/core/bitstreams/5c60f211-9c13-43ee-90dc-f5de2bc80743/content#55#6>). This indicates that the model itself is an approximation framework in which the proportionality coefficient is not fixed at 1.4 but varies with specific structural conditions. Setting the coefficient to 1 in this study is a simplification of this approximation framework, not a misinterpretation of Rogak's results.

(4) Subsequent observational validation indirectly supports this approximation: after adopting the simplification $R_{oe} \cong R_g$, FACM achieved significant improvements in visibility and surface shortwave radiation simulations, which demonstrates the reasonableness of this simplification at the level of results.

In summary, setting the coefficient to 1 is a methodological choice for clarity and focus. It does not negate the accuracy of the 1.4 coefficient under specific conditions. Rather, since the core of our derivation is to establish the power-law scaling relationship between D_f and the equivalent radii, any constant factor independent of N and D_f —whether 1.4 or 1—is absorbed into the prefactor and does not alter the power-law exponent $1/D_f$. The simplification to 1 serves to clearly highlight the dominant role of the fractal dimension D_f as the controlling physical parameter.

Revision:

Revise the Note for eq.14 to as follows:

Note: This approximation is based on the linear relationship between the projected area equivalent diameter and the radius of gyration of fractal aggregates. Rogak et al. (1993) demonstrated that for aggregates with fractal dimension $D_f \gtrsim 2$, the projected area equivalent diameter $d_p \approx 1.4R_g$, with the proportionality coefficient exhibiting weak dependence on cluster size and monomer number. In this study, to focus on the derivation of scaling exponents, the proportionality coefficient is simplified to 1. This simplification does not affect the core scaling behavior, because constant factors independent of N and D_f are absorbed into the prefactor and do not alter the power-law exponent. In fractal aerosol optics, the optical properties are known to be much less sensitive to the prefactor than to the fractal dimension (e.g., Liu et al., 2009, 2024; Romshoo et al., 2021). The reasonableness of this simplification is further supported by the subsequent observational validation of visibility and radiation simulations.

14. Line 342: I cannot follow the argument for why sedimentation calculations should use R_{oe} and why optical calculations should use R_{ae} , I would think the opposite is true.

Reply to the comments 14:

We sincerely thank the reviewer for carefully pointing out this error. You are entirely correct: in the description near Eq. (15), we reversed the equivalent radii that should be used for optical calculations and for sedimentation processes. The correct statement should be: **optical calculations must use the optical equivalent radius R_{oe} , while**

dynamical processes such as sedimentation must use the aerodynamic equivalent radius R_{ae} . The incorrect statement in the original manuscript was a typographical error. We have corrected it in the revised manuscript and have carefully checked the entire text paragraph by paragraph to ensure that all statements involving R_{oe} and R_{ae} are consistent with their definitions. In addition, we have checked all similar statements throughout the manuscript to ensure that no such reversal remains.

Revision in the main text:

Location: Section 2.2, near Eq. (15).

Revision: The erroneous sentence:

"optical calculations must use R_{ae} , while processes such as sedimentation must use R_{oe} "

has been changed to:

"optical calculations must use R_{oe} , while processes such as sedimentation must use R_{ae} ".

15. Line 364: What's FastJ?

Reply to the comments 15:

We thank the reviewer for this question. Fast-J is a photolysis scheme used in WRF-Chem, developed by (Wild, Zhu, & Prather, 2000) (full title: "Fast-J: Accurate simulation of in- and below-cloud photolysis in tropospheric chemical models"). It is designed to rapidly and accurately calculate photolysis rates in and below clouds in the troposphere, and it is one of the standard photolysis schemes widely used in WRF-Chem.

In this study, Fast-J is used because it is the default photolysis scheme coupled with the RADM2 gas-phase chemistry mechanism in WRF-Chem, and it is kept identical in both CTR and EXP. Fast-J requires aerosol optical parameters (such as the extinction coefficient) as input to compute actinic fluxes. Therefore, the modification of the aerosol optical equivalent radius in this study (R_{oe}) is passed to the photolysis rates through the optical calculations in Fast-J. In short, Fast-J is the photolysis module in WRF-Chem, used here to maintain the completeness of the model configuration and the normal operation of chemical processes. The core modification of this study (FACM) targets aerosol optical properties and affects photolysis only indirectly through Fast-J, without changing the direction of the conclusions.

16. Line 378: Perhaps "configure" should be "configuration"?

Reply to the comments 16:

Yes, "configure" has been corrected to "**configuration**".

17. Line 380: "validation"-> "validate"

Reply to the comments 17:

Yes, “validation” has been corrected to “**validate**”.

18. Table 1: For consistency, “photolysis” should be capitalized

Reply to the comments 18:

Yes, “photolysis” has been capitalized as “**Photolysis**” for consistency.

We thank the reviewer for these careful corrections.

19. Lines 437-438: Some more discussion about the justification for this fractal dimension should be provided: “based on estimates of aerosol composition ratios in the atmosphere” based on estimates, how? What estimates? Where were those estimates determined from? And what composition ratios? Also, is this a population fractal dimension (something like an effective fractal dimension), or are particles separated into groups, like organics and sulphates having $D_f=3$ and BC having $D_f=2.7$ after compaction? What about dust particles? A more detailed explanation of these points is necessary, because this is a central point in interpreting the results.

Reply to the comments 19:

We thank the reviewer for pointing out that the phrase “based on estimates of aerosol composition ratios in the atmosphere” was unclear, and for prompting us to provide a more detailed justification for $D_f = 2.7$. We fully agree that the choice of D_f is a central issue for interpreting the results, and we clarify it below.

(1) $D_f = 2.7$ is not derived from aerosol composition ratios; it is an empirical value based on sensitivity experiments and observational constraints.

The original phrase “based on estimates of aerosol composition ratios” was indeed too vague and could mislead readers into thinking that $D_f = 2.7$ was directly calculated from, for example, the mass ratios of black carbon, sulfate, and organics. This was not the case. We have corrected this statement in the revised manuscript.

(2) The rationale for $D_f = 2.7$ is that the average fractal dimension of aerosols in the real atmosphere should lie between 2.1 and 3.0.

Freshly formed, freely coagulated soot aggregates have a fractal dimension of about 2.1 (from DLA simulations and supported by numerous electron microscopy observations). Fully compact spheres without internal voids have $D_f = 3.0$. In the real atmosphere, aging processes—including condensation, coagulation, and hygroscopic compaction—drive D_f from 2.1 toward 3.0. The exact value depends on aerosol composition, aging degree, and humidity: a higher fraction of hygroscopic aerosols and higher humidity push D_f closer to 3, whereas a higher fraction of black carbon and drier conditions (which slow hygroscopic aging) push D_f closer to 2.1.

(3) This study adopts an effective fractal dimension for the overall aerosol population, rather than assigning D_f separately to different species.

The FACM scheme uses a single representative D_f to characterize the average fractal

properties of the entire aerosol population. It does not treat organics and sulfates as $D_f = 3$ and black carbon as $D_f = 2.7$ separately. This is because the optical module in WRF-Chem processes aerosols by size bins rather than by species-specific morphology, and it is difficult to separate the D_f of individual particles by species from observations. Therefore, using a single effective D_f is a reasonable simplification for the current parameterization.

(4) Treatment of dust aerosols.

Dust aerosols are indeed non-spherical, but their shapes are generally closer to ellipsoids or irregular compact bodies rather than fractal aggregates; their D_f should therefore be close to 3. The FACM scheme mainly targets combustion-derived fractal aggregates (e.g., black carbon and soot), and its applicability to dust is limited. In the East Asian sensitivity experiments, we also noticed that the impact of FACM over dust-dominated regions differs from that over combustion-dominated regions, indicating that the applicable scope of FACM needs further definition. This also shows that the single effective D_f currently adopted is a reasonable approximation for combustion-dominated regions.

(5) The final choice of $D_f = 2.7$ is based on the sensitivity experiments in Supplementary Material S1.

We compared AOD and near-surface visibility simulations for several values of D_f , including 3.0, 2.7, 2.4, and 2.1. The results show that when D_f is reduced from 2.7 to 2.4 or 2.1, the AOD increases significantly and the visibility decreases sharply, far exceeding the range supported by observations. In contrast, $D_f = 2.7$ gives results closest to observations. Therefore, $D_f = 2.7$ is an empirical value determined under observational constraints and through repeated tests, rather than a precise derivation from aerosol composition ratios. See Supplementary Material S1 for details.

So, we revised the following paragraph:

For the numerical simulations in this study, an average fractal dimension of approximately 2.7 was selected, based on estimates of aerosol composition ratios in the atmosphere. But it is important to acknowledge that using the fixed fractal dimension ($D_f \approx 2.7$) is a simplification. This value was chosen as a representative estimate for the overall aerosol mixture during winter haze events in the North China Plain, based on considerations of aging processes—including compaction due to coagulation, condensation, and hygroscopic growth—that tend to increase D_f over time. While this approximation is more physically realistic than the traditional assumption of compact spheres ($D_f = 3$), it does not capture the full range of D_f variability observed under different conditions.

to be:

For the numerical simulations in this study, an average fractal dimension of approximately 2.7 was adopted. **This value should be understood as an effective fractal dimension representing the overall aerosol population, rather than a value derived from a specific aerosol composition ratio. In the real atmosphere, the average fractal dimension of aerosols is expected to lie between 2.1 (freshly formed, freely coagulated**

soot aggregates) and 3.0 (fully compact spheres). Aging processes, including compaction due to coagulation, condensation, and hygroscopic growth, tend to increase D_f toward 3, with the exact value depending on aerosol composition, aging degree, and humidity. The choice of $D_f=2.7$ was determined through sensitivity experiments comparing $D_f=3.0, 2.7, 2.4$, and 2.1 (see Supplementary Material S1), in which $D_f=2.7$ gave results closest to observations. While this fixed value is a simplification that does not capture the full range of D_f variability, it is more physically realistic than the traditional assumption of compact spheres ($D_f=3$).

20. Lines 447-450: There are several other published works that discuss this process, besides the Wang and the Bai studies. Some citations would be useful.

Reply to the comments 20:

We thank the reviewer for this helpful suggestion. We agree that additional citations are needed to support the statement that the restructuring of fractal soot aggregates under humid conditions has been documented in both laboratory experiments and field observations. In the revised manuscript, we have expanded the reference list to include several additional published works, as follows:

Zelenay et al. (2011) investigated the chemical and physical properties of combustion particles from diesel vehicles and a wood stove before and after photochemical aging. They found that hygroscopic behavior relates well with chemical signatures, and that higher amounts of carboxylic and phenolic carbon lead to higher water uptake by aggregated particles, resulting in noticeable restructuring upon humidification.

Zhang et al. (2008) showed that soot particles exposed to subsaturated sulfuric acid vapor exhibit a marked change in morphology, characterized by a decreased mobility-based diameter but an increased fractal dimension and effective density, and that coating with sulfuric acid and subsequent hygroscopic growth enhance the optical properties of soot aerosols.

Khalizov et al. (2013) investigated the role of OH-initiated oxidation of isoprene in the aging of combustion soot and found that the restructuring of coated aggregates is caused by surface tension forces exerted by the liquid coating, with the mass fractal dimension of soot initially decreasing and then increasing as the coating thickness grows.

Li et al. (2023) studied the restructuring of diesel soot particles in a simulated atmospheric environment and observed that the fractal dimension tends to increase during the aging process for both low- and high-load soot, with the low-load soot showing a higher aging degree and faster aging speed.

Beeler et al. (2025) developed a framework for predicting the size and fractal dimension of compacting soot particles as a function of coating volume ratio, confirming that compaction of fractal BC aggregates is caused by condensation of non-BC species and that the fractal dimension evolves throughout compaction.

Li et al. (2024) provided a comprehensive review of the microphysical properties of atmospheric soot and organic particles, emphasizing that fractal dimension is a commonly employed quantitative metric for characterizing soot morphologies and their modifications in relation to aging factors such as internal mixing state, core-shell structures, phase, and composition heterogeneity.

These studies collectively confirm that the transformation of fractal soot aggregates toward more compact morphologies under humid and aged conditions has been documented across a range of laboratory and field investigations.

Revision in the main text:

The sentence:

These transformations have been documented in both laboratory experiments and field observations (Y. Wang, et al., 2017; Bai, et al., 2018).

has been replaced by:

These transformations have been documented in both laboratory experiments and field observations (Bai et al., 2018; Beeler, Corbin, Sipkens, & Fierce, 2025; Khalizov et al., 2013; D. Li, Wei, Chen, Wang, & Wang, 2023; W. Li et al., 2024; Wang et al., 2017; Zelenay et al., 2011; Zhang et al., 2008)

21. Lines 628-630: “It is important to clarify that the FACM correction applies to all aerosol species exhibiting fractal morphology (not only black carbon)”;

what other fractals are they? Can the authors be more explicit?

Reply to the comments 21:

We thank the reviewer for prompting us to more precisely define the applicability of the FACM correction. In the revised manuscript, we clarify that the FACM correction is, in principle, applicable to all aerosol species that form through free coagulation and persist in the atmosphere long enough to develop fractal morphology before being compacted by aging, not only black carbon.

Observational evidence has confirmed fractal-like structures in several non-black-carbon aerosol types:

- a) **Biomass burning tar balls:** In wildfire smoke plumes in New Mexico, about 27% (by number) of tar balls were observed as aggregates with fractal-like structures, following scale-invariant power laws similar to those of soot particles (Giroto et al., 2018). Aggregation was also found to significantly affect their optical properties and thus their climate effects.
- b) **Dust aggregates:** Using scanning electron microscopy image analysis, dust particles collected during a severe dust storm in Israel were successfully separated into two distinct particle groups based on their fractal dimensions, confirming the fractal characteristics of dust aggregates (Koren, Ganor, & Joseph, 2001).
- c) **Secondary organic aerosols (SOA) from photooxidation:** A study using an

environmental chamber to investigate SOA formation from diesel exhaust photooxidation measured the effective density and fractal dimension of particles, showing that SOA forms on fractal particle surfaces and that the fractal dimension is a key parameter describing this process (Nakao, Shrivastava, Nguyen, Jung, & Cocker, 2011).

Therefore, the FACM parameterization is not limited to any specific species; its optical correction is based on the geometric parameter D_f and can in principle be applied to all aerosols with fractal morphology. In most cases of interest, however, combustion-derived fractal aggregates (e.g., soot, black carbon, tar balls) dominate in anthropogenically polluted regions, where observational evidence is also most abundant. The explicit mention of "other" cases beyond soot is included for theoretical rigor. We have added these references and specified these "other" cases in the revised manuscript.

In Section 4.4 (begin at Line 650)

the original sentence: "It is important to clarify that the FACM correction applies to all aerosol species exhibiting fractal morphology (not only black carbon)."

has been replaced by:

"It is important to clarify that the FACM correction is, in principle, applicable to all aerosol species that exhibit fractal morphology, not only black carbon. Observational evidence has confirmed fractal-like structures in biomass burning tar balls (Giotto et al., 2018), dust aggregates (Koren et al., 2001), and secondary organic aerosols (Nakao et al., 2011). In this study, the FACM parameterization is applied to all size bins that contain fractal aggregates, with a focus on combustion-derived aerosols (e.g., soot, black carbon, tar balls) that dominate in anthropogenic pollution regions."

Reference in this response to comments:

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Supplementary Materials for “Fractal Geometry of Aerosol Particles and Its Impact on Atmospheric Optical Properties: Development and Validation of the Fractal Aerosol Cluster Model in a Heavy Haze Event”

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S1 Sensitivity experiments of fractal dimension over North China

S1.1 Experimental setup

Based on the North China case in the main text (24–27 November 2018), near-surface extinction coefficients were calculated at three selected times (25 November 00:00 and 12:00, and 26 November 00:00) for $D_f = 3.0, 2.7, 2.4$, and 2.1 , respectively, and the corresponding AOD and VIS were further derived.

Among these, $D_f = 2.4$ and $D_f = 2.1$ are the sensitivity experiment settings for D_f performed in response to the commenter's request. They can also be regarded as extreme hypothetical scenarios in which the aging mechanism of black carbon aerosols is abruptly weakened.

S1.2 Sensitivity results of AOD

The AOD increases significantly as D_f decreases. The AOD at $D_f = 2.1$ far exceeds the reasonable range of observations, demonstrating that D_f is a key parameter controlling AOD.

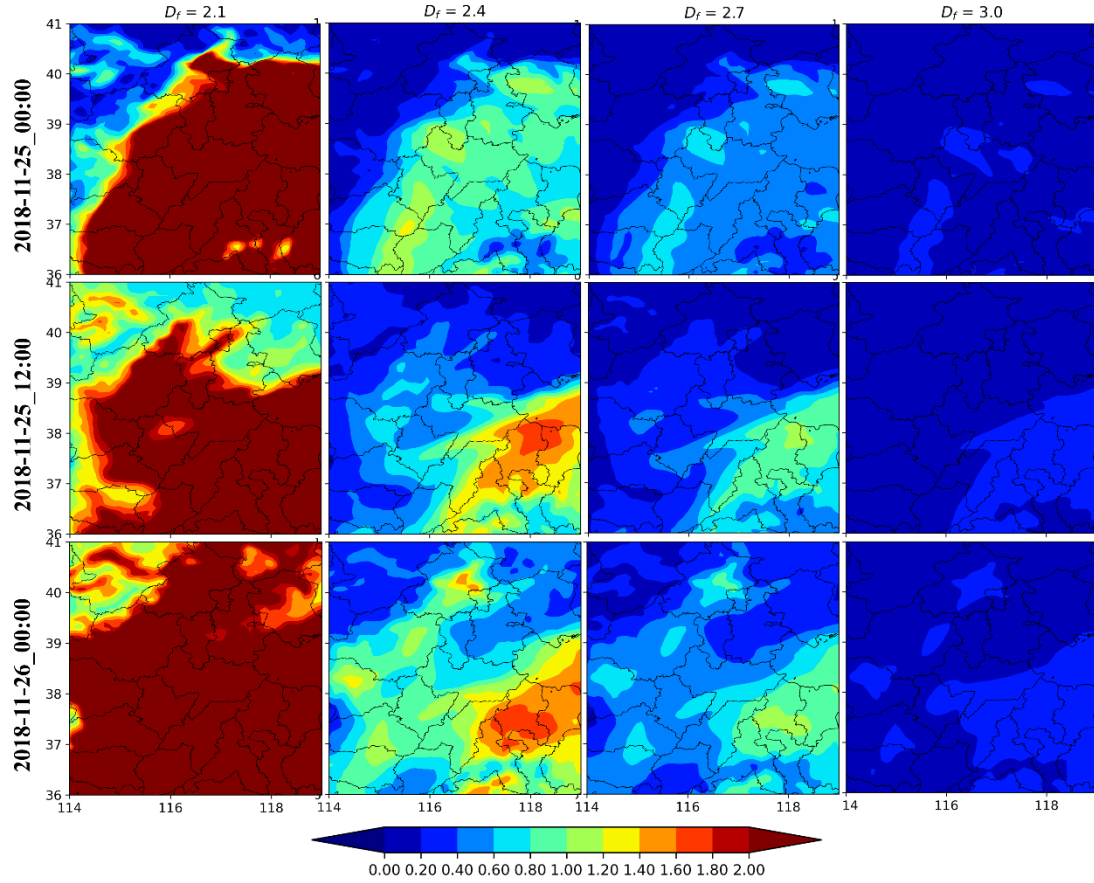


Figure S1: Spatial distribution of AOD over North China at three selected times for $D_f = 3.0, 2.7, 2.4, 2.1$.

S1.3 Sensitivity results of VIS

The VIS decreases sharply as D_f decreases. The VIS at $D_f = 2.1$ is far lower than the observations, demonstrating the strong sensitivity of visibility simulations to D_f . Meanwhile, the results for $D_f = 2.7$ agree most closely with the observations, indicating that it is a reasonable choice.

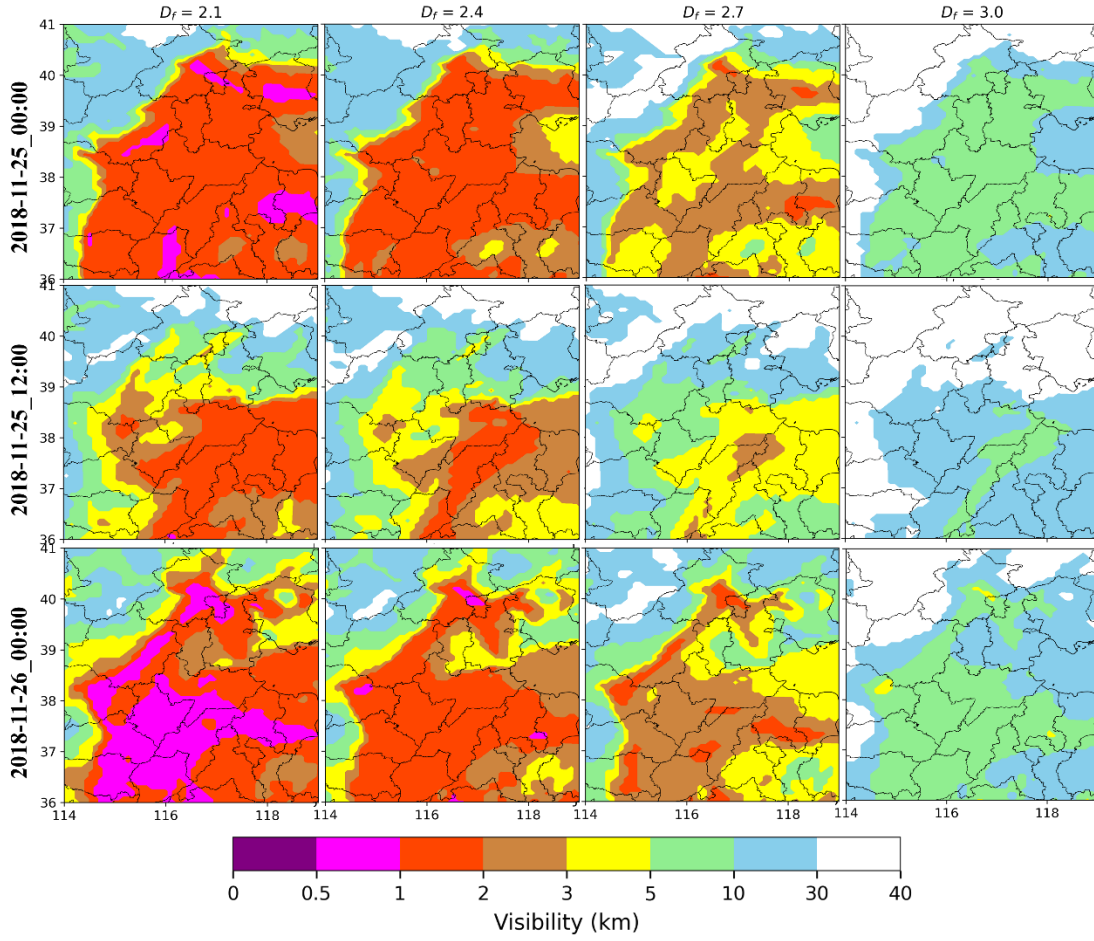


Figure S2: Spatial distribution of near-surface VIS over North China at three selected times for different D_f values, using the same discrete color scale as in the main text.

S1.4 Summary

The main findings of the sensitivity experiments are summarized as follows: (1) D_f exerts a strong control on both AOD and VIS; (2) $D_f = 2.1$ represents unaged fresh soot, and aging processes in the real atmosphere shift it toward 3; (3) $D_f = 2.7$ is a reasonable approximation after aging for the winter haze events over the North China Plain.

S2 Sensitivity experiments of FACM over East Asia

S2.1 Brief description of the model setup

To evaluate the applicability of FACM at a larger spatial scale, an additional set of sensitivity experiments was conducted over East Asia. The simulation domain covers the East Asian continent and adjacent seas, using a single nested grid with a horizontal resolution of 25 km, for the period from 1 to 31 December 2019.

The experimental design follows that of the North China case in the main text, consisting of a control group (CTR, original WRF-Chem) and an experimental group (EXP, WRF-Chem coupled with FACM). Both experiments use identical meteorological initial and boundary conditions and identical physical and chemical parameterization schemes; only the treatment of the equivalent radius in the aerosol optical module differs. The meteorological initial and boundary conditions are taken from the NCEP FNL reanalysis data; anthropogenic emissions are from the MEIC inventory, biogenic emissions from MEGAN, and biomass burning emissions from FINN, while the chemical initial and boundary fields are from CAM-chem output. The physical and chemical schemes include: RRTMG longwave/shortwave radiation, RADM2/SORGAM aerosol chemistry, RADM2 gas-phase chemistry, Fast-J photolysis, Purdue Lin microphysics, Grell 3D cumulus, MYNN2 boundary layer, and Noah land surface processes. It should be noted that this part focuses on evaluating the qualitative impact of FACM at the East Asian scale, and no station-by-station quantitative validation was performed over East Asia; the quantitative validation has already been completed for the North China, Tianjin, and Shenyang cases in the main text.

S2.2 AOD results over East Asia

The high-AOD regions are mainly located over the Central Asian deserts and northern India. The Kyzylkum, Karakum, and Taklamakan deserts in Central Asia are the main sources of dust aerosols; the high AOD over northern India is associated with substantial anthropogenic combustion emissions (black carbon and soot) as well as mixing with dust from the Thar Desert. The AOD simulated by EXP over Central Asia and northern India is generally greater than 0.4, locally approaching 1.0, whereas the maximum AOD in CTR does not exceed 0.5. Over other parts of the East Asian continent, the AOD in EXP is also about 0.1 higher than in CTR, and a local high-AOD feature appears in the South China–Taiwan region. The difference map indicates that CTR overall underestimates AOD, especially over northern India and Central Asia, where the AOD in EXP is about 100%–150% higher than in CTR. This suggests that FACM is more sensitive to the extinction enhancement of non-spherical aerosols with pronounced fractal characteristics, such as dust and carbonaceous aerosols.

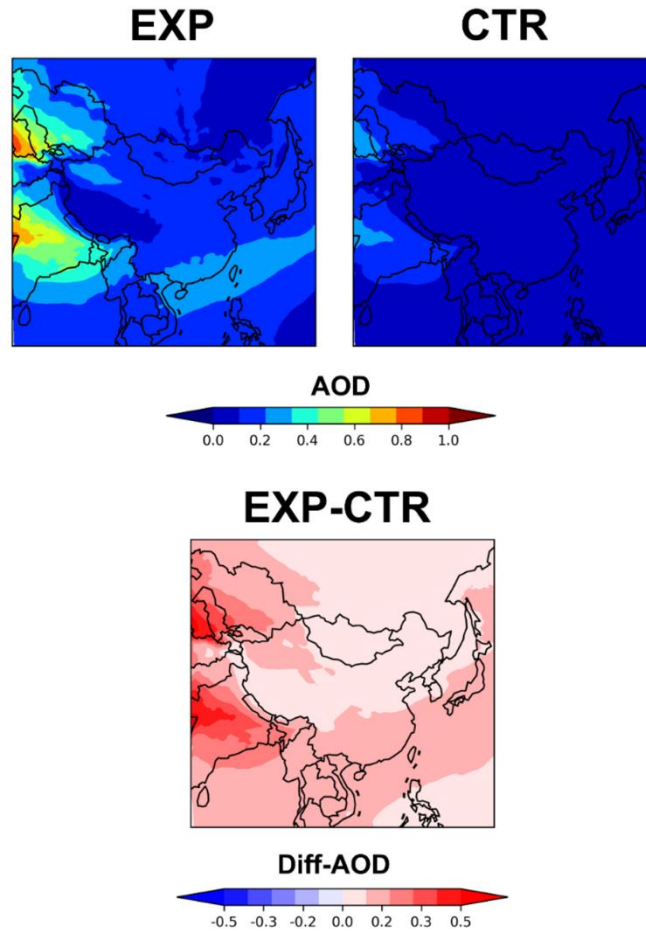


Figure S3: Simulated average AOD over East Asia in December 2019 from EXP and CTR, and their difference (EXP–CTR).

S2.3 SWD and Qcl results over East Asia

The SWD differences between EXP and CTR are mainly concentrated over northern India, the Central Asian deserts, and the South China–Taiwan region, reaching about 20 W m^{-2} over northern India. These regions coincide closely with the high-AOD areas, indicating that enhanced aerosol extinction directly reduces the shortwave radiation reaching the surface. The SWD difference peak over the South China – Taiwan region overlaps with the positive Qcl difference area, suggesting that the reduced SWD in this region may be partly related to increased cloud water. When fractal aerosols act as cloud condensation nuclei, their complex aggregate structures may alter the initial morphology and growth process of cloud droplets, thereby affecting cloud amount and radiative transfer. Over oceanic regions, both AOD and SWD differences are small, consistent with the fact that marine aerosols tend to become spherical after hygroscopic growth and thus exhibit weak fractal effects.

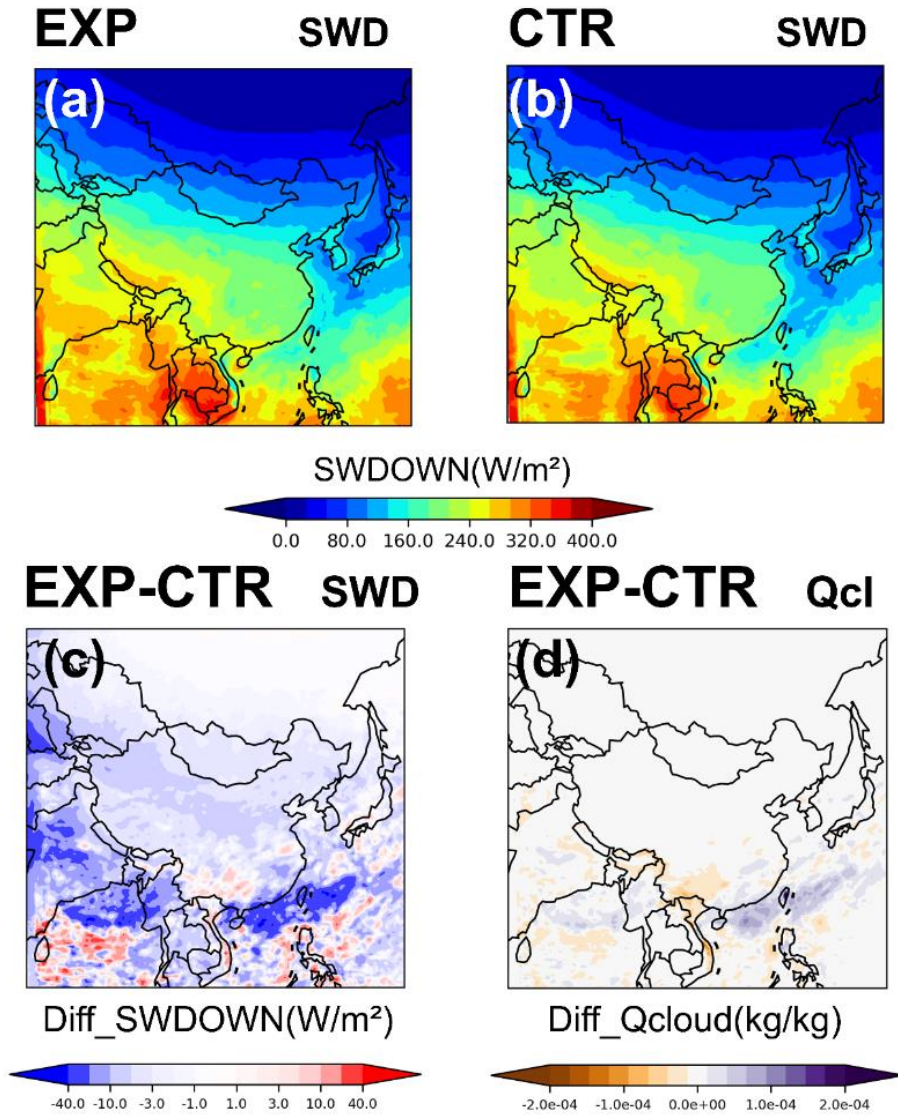


Figure S4 Simulated average SWD and Qcl over East Asia from EXP and CTR, and their differences.

S2.4 TSK results over East Asia

The TSK simulated by EXP is overall lower than that by CTR, especially over the Central Asia–India region and near Sakhalin Island in Outer Northeast Asia, with an average decrease of more than 0.5 °C over Central Asia and India; over the ocean, the difference between the two is close to zero.

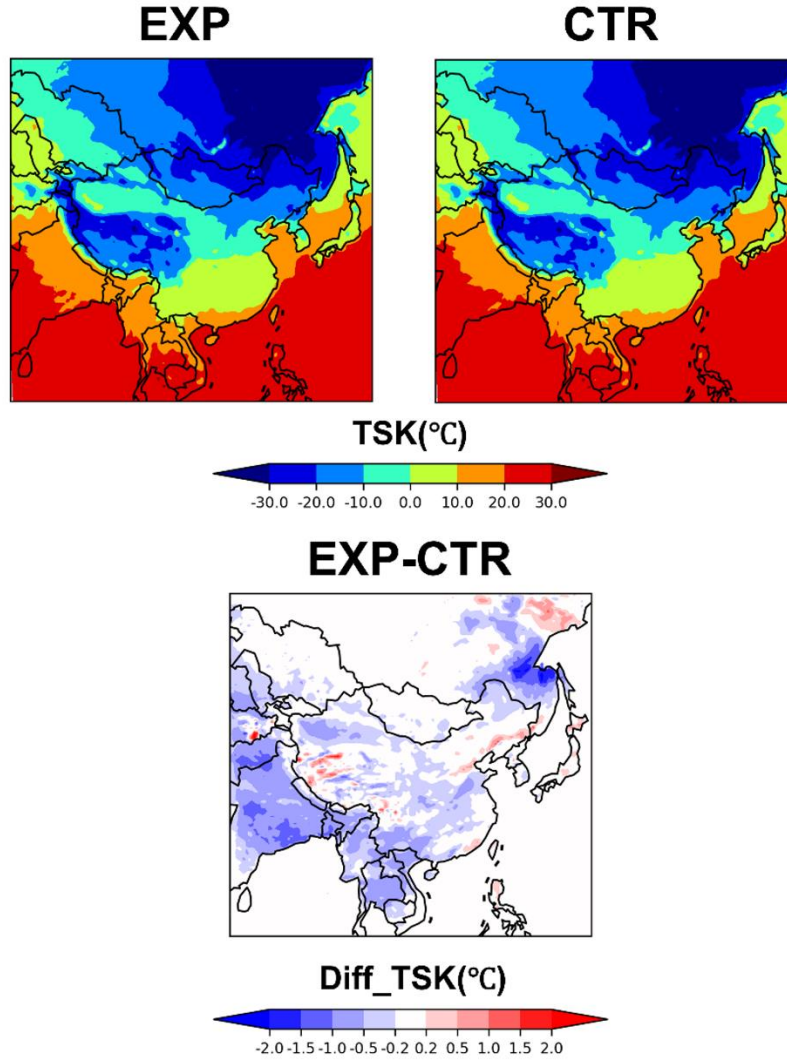


Figure S5: Simulated average TSK over East Asia from EXP and CTR, and their difference.

S2.5 AOD–SWD correlation analysis

After screening for clear-sky daytime conditions with $\text{AOD} > 0.2$, i.e., aerosol-dominated conditions, ΔAOD and ΔSWD are mainly negatively correlated, approaching -1.0 over parts of Central Asia and northern India. This indicates that fractal aerosols intensify the attenuation of shortwave radiation by enhancing scattering and absorption.

S2.6 AOD–TSK correlation analysis

The correlation between ΔAOD and ΔTSK exhibits regional differences: over the Central Asian deserts, where scattering dust dominates, an increase in AOD leads to surface cooling (negative correlation); over northern India and North China, where absorbing aerosols (such as black carbon) account for a relatively high proportion, the fractal characteristics enhance atmospheric heating and indirectly lead to surface warming (positive correlation) by suppressing convection or increasing downward

longwave radiation. This indicates that the radiative effect of fractal aerosols depends on regional aerosol chemical composition and geographical environment.

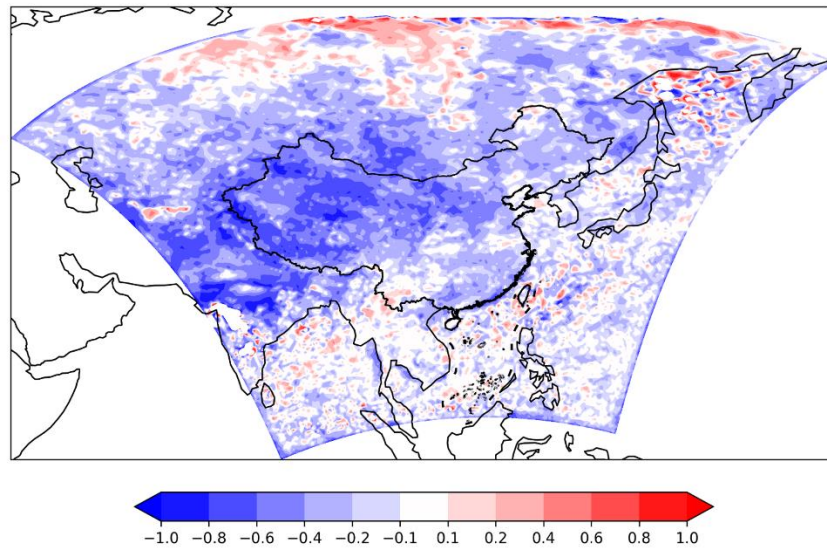


Figure S6: Spatial distribution of the correlation between the AOD difference and the SWD difference from EXP and CTR.

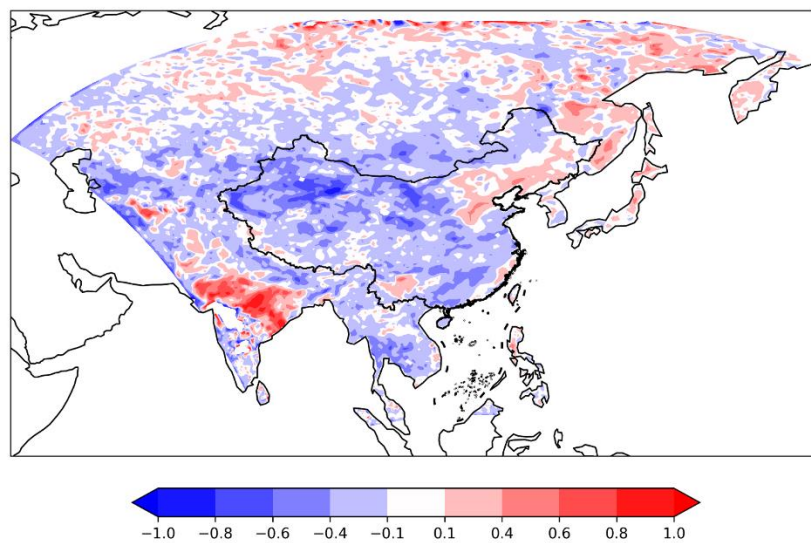


Figure S7: Spatial distribution of the correlation between the AOD difference and the TSK difference from EXP and CTR.

S2.7 Summary

The East Asian sensitivity experiments show that FACM can still capture the significant effects of fractal aerosols on AOD, SWD, and TSK at a larger spatial scale, verifying its transferability and its potential for improving aerosol radiative forcing estimates in GCMs.

S3: Diffusion-Limited Aggregation (DLA) algorithm along with MATLAB scripts for model visualization (Text S1);

The DLA algorithm employed in this study consists of three main components. The first part is the main program body, the function `fractal_growth_rad`, which simulates the complex and irregular shapes of aerosol clusters as described in section 2.1 of the paper. Additionally, the algorithm calculates the fractal dimension and invokes other functions for the purpose of visualization.

The main part of the DLA algorithm

```
function structure = fractal_growth_rad(N)
    % The size of three-dimensional space
    gridSize = 100;
    % Initialize the three-dimensional space
    structure = zeros(gridSize, gridSize, gridSize);
    origin = [ceil(gridSize/2), ceil(gridSize/2), ceil(gridSize/2)];

    % Place a fixed particle at the origin at the beginning
    structure(origin(1), origin(2), origin(3)) = 1;

    % Define the neighborhood: Use a spherical neighborhood with a
    radius of 1.5
    neighborOffsets = create_spherical_neighbors(1.5);

    % Initialize the plot
    figure;
    axis equal;
    hold on;
    view(3);
    xlim([0 gridSize]);
    ylim([0 gridSize]);
    zlim([0 gridSize]);
    title(['Dynamic Fractal Aerosol Cluster Growth.'
    N=', num2str(N)]);

    % Add global light sources
    light('Position', [1 1 1], 'Style', 'infinite');
    light('Position', [-1 -1 -1], 'Style', 'infinite');

    draw_sphere(origin(1), origin(2), origin(3), 1);
```

```

% Random walk and build the structure
for i = 1:N
    % Randomly generate a particle initially
    particlePos = origin + randi([15, gridSize/2], 1, 3).*sign(rand(1, 3) - 0.5);

    while true
        % Randomly walk to an adjacent grid point
        step = neighborOffsets(randi(size(neighborOffsets, 1)), :);
        particlePos = particlePos + step;

        % If it goes out of bounds, randomly generate the particle again
        if any(particlePos <= 1) || any(particlePos >= gridSize)
            particlePos = origin + randi([15, gridSize/2], 1, 3).*sign(rand(1, 3) - 0.5);
            continue;
        end

        % Check if it is near an existing structure
        for j = 1:size(neighborOffsets, 1)
            neighborPos = particlePos + neighborOffsets(j, :);
            if structure(neighborPos(1), neighborPos(2), neighborPos(3)) == 1
                % Calculate the distance to the origin
                distToOrigin = norm(particlePos - origin);

                % Calculate a fixed probability based on the distance
                fixationProb = 0.01 + 3.99 * (distToOrigin / (gridSize/2));

                % particle fixed?
                if rand < fixationProb
                    structure(particlePos(1), particlePos(2), particlePos(3)) = 1;
                    % Dynamically draw the newly grown particle
                    draw_sphere(particlePos(1), particlePos(2), particlePos(3), 1.0);
                    drawnow;
                    pause(0.001);
                    break;
                end
            end
        end
    end
end

```

```

        end
    end
    if      structure(particlePos(1),      particlePos(2),
particlePos(3)) == 1
        break; % The particle is fixed, stop this random walk
    end
end
end

% Calculate the fractal dimension - Box Counting Method
boxCounts = [];
boxSizes = 2.^(0:5);
for boxSize = boxSizes
    numBoxes = ceil(gridSize / boxSize);
    boxCount = 0;
    for x = 1:numBoxes
        for y = 1:numBoxes
            for z = 1:numBoxes
                xRange = (x-1)*boxSize+1:min(x*boxSize, gridSize);
                yRange = (y-1)*boxSize+1:min(y*boxSize, gridSize);
                zRange = (z-1)*boxSize+1:min(z*boxSize, gridSize);
                if any(any(any(structure(xRange, yRange, zRange))))
                    boxCount = boxCount + 1;
                end
            end
        end
    end
    boxCounts = [boxCounts, boxCount];
end

% Fit the relationship between box size and box count to calculate
the fractal dimension
logBoxSizes = log( 1./boxSizes);
logBoxCounts = log(boxCounts);
fractalDim = polyfit(logBoxSizes, logBoxCounts, 1);

% Display the fractal dimension
fprintf('Fractal Dimension: %f\n', fractalDim(1));
disp(['Fractal Dimension=', num2str(fractalDim(1))])
% Plot the box counting graph
figure;
scatter(logBoxSizes, logBoxCounts, 'filled');
hold on;

```

```

% Add the fitted line
fittedLine = polyval(fractalDim, logBoxSizes);
plot(logBoxSizes, fittedLine, 'r--', 'LineWidth', 2);

xlabel('log(1/Box Size)');
ylabel('log(Box Count)');
title('Box Counting Method: Fractal Dimension');
hold off;
end

```

Meanwhile, to ensure the normal operation of the main program, we have also constructed a function `neighborOffsets`. Its purpose is to create a spherical neighborhood.

Create a spherical neighborhood.

```

function neighborOffsets = create_spherical_neighbors(radius)
    [x, y, z] = ndgrid(-ceil(radius):ceil(radius), -
        ceil(radius):ceil(radius), -ceil(radius):ceil(radius));
    distances = sqrt(x.^2 + y.^2 + z.^2);

    % Select points within the spherical radius, but not including
    the origin itself.
    valid = (distances <= radius) & (distances > 0);
    neighborOffsets = [x(valid), y(valid), z(valid)];
end

```

Furthermore, we use the function `draw_sphere` to plot the clusters, making them more visually intuitive.

Plotting the spatial structure of fractal clusters.

```

function draw_sphere(x, y, z, r)
    % Draw a sphere with a 3D grayscale gradient effect at the
    specified position with radius r
    [sx, sy, sz] = sphere(50);
    sphereX = r * sx + x;
    sphereY = r * sy + y;
    sphereZ = r * sz + z;
    colormap(gray);
    % Generate a grayscale color matrix, using normalized values of
    the 3D coordinates for the grayscale gradient
    C = sqrt(sx.^2 + sy.^2 + sz.^2); % Generate grayscale using
    normalized distance from the center of the sphere on the surface
    % Plot the sphere with a grayscale gradient
    surf(sphereX, sphereY, sphereZ, C, 'FaceColor', 'interp',
        'EdgeColor', 'none', 'FaceLighting', 'gouraud');

```

end

S4. Time series plots of visibility simulations and observations at different stations (Figure S8)

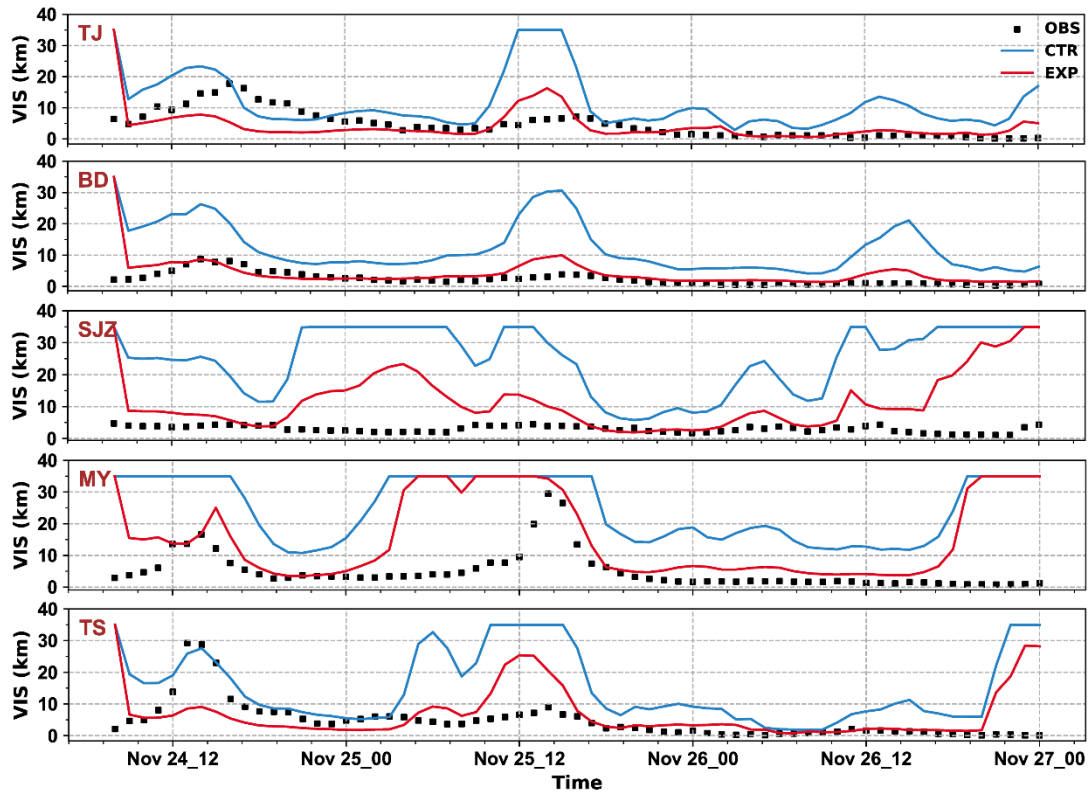


Figure S8. The AV timeseries at station TJ, BD, SJZ, MY and TS: the black square markers (OBS) represent the observation; the blue line (CTR) stands for other by CTR and the red line (EXP) by EXP simulation.

S5. Dependence of the optical-to-aerodynamic radius ratio on fractal dimension and cluster size

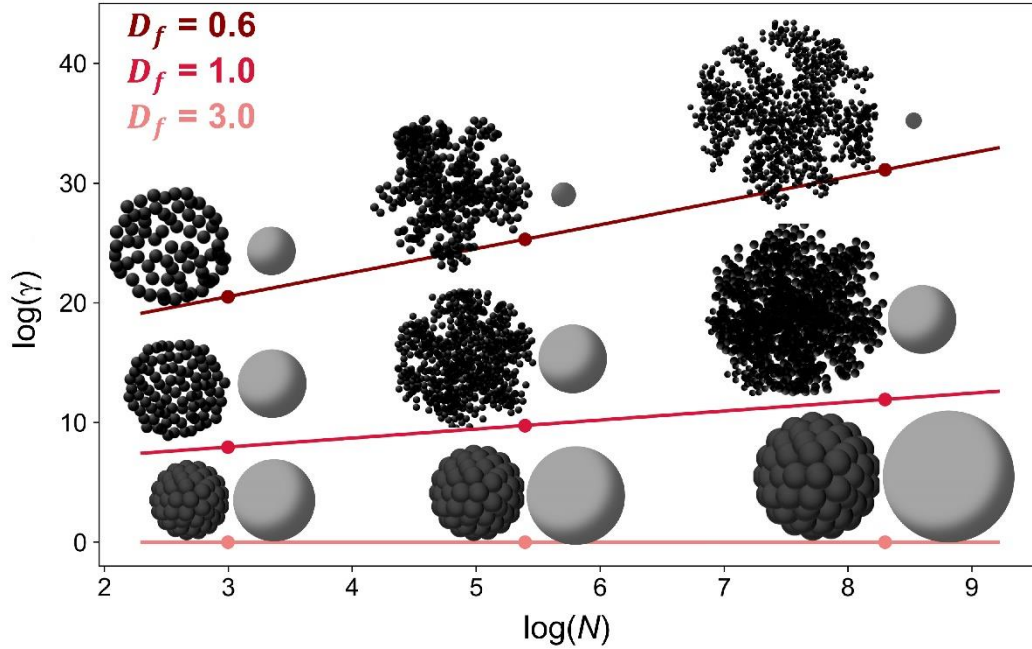


Figure S9. monomer number $\log(N_s)$ and fractal dimension D_f versus $\log(\gamma)$. The different color intensity of solid lines represents different fractal dimensions D_f . The black markers and their sizes represent for the aerosol cluster and its optical equivalent radius. While the gray markers represent for those aerodynamical spherical equivalent radii.

According to Fig. S2 (Figure 2 in the previous draft) we show the most intrinsically different feature of fractal clusters compared to previous aerosol particles with a default dimension of 3: i.e., as the aerosol size (N) grows, the optically equivalent radius of the clusters grows faster than the aerodynamically equivalent radius and the ratio γ of the two is not a linear factor, but rather there is a power-law difference, which is related to the cluster's fractal geometry feature: the fractal dimension of the cluster: D_f . The further D_f deviates from 3, the larger the value of $\log \gamma$, i.e., a cluster with a smaller aerodynamic equivalent radius also has a stronger optical equivalent radius, which results in a stronger atmospheric scattering signature.

Exceptionally, when D_f is smaller than 1 (a situation that holds only in theoretical derivations, and no such exaggerated examples seem to have been confirmed to have been found in the current sampling observations), it is even possible to have a situation in which the optically equivalent radius of a cluster grows rapidly as the monomers continue to coalesce while the aerodynamically equivalent radius decreases at the same time.

The relevant theoretical derivation is in Sec.2.2 of the manuscript.