

Reply on CC1

This is an interesting and potentially useful study. The consideration of aerosol fractal morphology provides a physically meaningful extension to the conventional spherical-particle assumption, and the implementation in WRF-Chem makes the work relevant to both air-quality and aerosol–climate modeling. The manuscript is generally complete, and the main conclusions are reasonably supported by the numerical experiments and observations. I only have a few minor comments that may help improve the clarity and robustness of the paper.

1. The choice of the fractal dimension could be explained more clearly. The DLA simulation gives $D_f \approx 2.1$, while $D_f = 2.7$ is adopted in WRF-Chem, and the manuscript also notes observational values around 1.8–2.2 for soot particles. Although the authors provide reasonable explanations related to aerosol aging, a short sensitivity test using several D_f values, or at least a quantitative discussion of how the main results depend on this assumption, would strengthen the paper.
2. Please check the description of optical and aerodynamic equivalent radii around Eq. (15). The manuscript states that “optical calculations must use R_{ae} , while processes such as sedimentation must use R_{oe} .” This appears to be reversed based on the definitions provided earlier and should be carefully checked and corrected.
3. I suggest adding some discussion about the transferability of this FACM module to other models, especially the potential usage in GCMs to improve aerosol forcing estimation,

Our Response to Li Jing:

Reply to Comment 1

We greatly appreciate the reviewer’s constructive comment. We fully agree that a quantitative assessment of the sensitivity to the adopted value of the fractal dimension D_f would significantly strengthen the robustness of the conclusions of this paper. To this end, we have added a set of sensitivity experiments for D_f , which are presented in detail in the Supplementary Material.

1. Design of the sensitivity experiments

Based on the North China case simulation in the main text (24–27 November 2018), we designed four experiments for three selected times (25 November 00:00 and 12:00, and 26 November 00:00), with $D_f = 3.0$ (CTR, spherical assumption), $D_f = 2.7$ (EXP, the experimental setup in this paper), $D_f = 2.4$, and $D_f = 2.1$, respectively. The AOD and near-surface horizontal visibility (VIS) were calculated for the four experiments, and the results are shown in the Supplementary Material (Figures S1–S2).

2. Main findings

The sensitivity experiments reveal a key conclusion: even a small change in D_f has an extremely significant impact on AOD and VIS. When D_f is further reduced from 2.7 to 2.4 and 2.1, the AOD increases significantly, the VIS decreases sharply, and the magnitude of these changes far exceeds the realistic range that observations can support. This result is scientifically significant in two respects:

First, it strongly demonstrates that the fractal morphological characteristics of aerosol clusters have a high sensitivity to macroscopic atmospheric optical properties and are a core factor worthy of in-depth study. This is fully consistent with the core argument of this paper, that is, D_f is a key parameter controlling the extinction capability of aerosols. Second, it indicates that $D_f = 2.1$, taken as an extreme assumption of purely free coagulation growth, is theoretically valid but cannot represent the average aerosol state after aging processes in the real atmosphere.

3. Physical meaning and limitations of $D_f = 2.1$

As described in Section 2 of the main text, the fractal dimension of the clusters simulated by the DLA algorithm is approximately 2.1, which corresponds to the structural characteristics of freshly generated soot aggregates that have not undergone any external compaction and are formed through free coagulation. Numerous electron microscopy observations have also confirmed that the D_f of fresh soot collected at tunnel stations is about 1.8–2.0, while the D_f of aged soot at background stations can rise above 2.0 (Wang et al., 2017). However, aerosols in the real atmosphere undergo a series of rapid aging processes after emission:

(1) Condensation aging: Sulfuric acid, nitric acid, and organic vapors condense on the soot surface and fill the internal voids of the aggregates. In the Beijing area, the aging timescale dominated by condensation in the daytime surface layer can be as short as less than 2 hours (Peng et al., 2016).

(2) Coagulation aging: Soot coagulates with soluble aerosols such as sulfates and nitrates, further altering the aggregate structure. Although the timescale of coagulation aging at night is longer than that of condensation, it can still be completed within several hours to one day (Chen et al., 2017; Moteki et al., 2007).

(3) Hygroscopic compaction: After the coating absorbs water, the surface tension of liquid water forces the aggregates to restructure toward a denser and more nearly spherical morphology. Experiments show that after sulfuric acid coating and exposure to 80% relative humidity, the scattering of soot can be enhanced by about a factor of 10 and the absorption by nearly a factor of 2 (Zhang et al., 2008). The timescale for hygroscopic restructuring and compaction is estimated to be less than 0.7 days (Mikhailov, Vlasenko, Podgorny, Ramanathan, & Corrigan, 2006).

The combined effect of these aging processes is to shift D_f from about 2.1 in the fresh state toward 3. Therefore, the average fractal dimension of aerosols in the real atmosphere should logically lie between 2.1 and 3. The specific value exhibits pronounced spatiotemporal variability depending on emission sources, chemical conditions, meteorological factors (especially humidity), and other factors.

4. Basis for choosing $D_f = 2.7$

In the simulation of the winter haze event over the North China Plain, we adopt $D_f = 2.7$ based on the following considerations: this value lies between that of fresh soot ($D_f \approx 2.1$) and that of a fully compact sphere ($D_f = 3$), representing a mixed aerosol population that has undergone partial aging and hygroscopic compaction. Although the mass fraction of fresh black carbon in PM_{2.5} is typically only 5–10%, after several hours to days of aging, the mass fraction of black carbon-containing aerosols in polluted urban atmospheres can reach 32–52%, and most of them are in a thickly coated state (with a coating thickness parameter D_p/D_c of about 1.6–2.2), indicating that aging processes are highly efficient in polluted atmospheres (Wu et al., 2019). Therefore, $D_f = 2.7$ is a relatively conservative engineering approximation for the overall aging degree of aerosols during winter haze events over the North China Plain—if a smaller D_f were adopted, the fractal effect would only be stronger and the improvement from FACM would be even more pronounced, without changing the direction of the conclusions.

5. Summary

The sensitivity experiments show that: (1) D_f is a highly sensitive parameter controlling aerosol optical properties; (2) $D_f = 2.1$ represents freshly generated soot formed by free growth, whereas aging processes in the real atmosphere drive it toward 3; and (3) $D_f = 2.7$ is a reasonable value based on observational constraints and multiple tests, representing the aged mixed aerosol state during winter haze events over the North China Plain. The cases with $D_f = 2.1$ and 2.4 can be understood as extreme hypothetical scenarios—namely, assuming that the aging mechanism of fractal aerosols suddenly disappears—and their results demonstrate how exaggerated the changes in atmospheric optical properties would be under such an assumption, thereby further highlighting the importance of accounting for realistic aging processes. We thank the reviewer for prompting us to add this important sensitivity analysis.

Reply to Comment 2

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} ".

Reply to Comment 3: Transferability of the FACM module and its prospects for application in GCMs

We greatly appreciate the reviewer's forward-looking suggestion. We fully agree that the transferability of FACM and its potential for improving aerosol radiative forcing estimates in global climate models (GCMs) is an important direction for extending this study.

We would like to make the following points:

First, FACM has good portability. The scheme relies on relatively few parameters, mainly including the fractal dimension D_f , the monomer radius a , the scaling relationship between the radius of gyration R_g and the number of monomers N , and the resulting optical equivalent radius R_{oe} and aerodynamic equivalent radius R_{ae} . These parameters are available or can be diagnosed in most aerosol modules, so FACM can be readily embedded into the aerosol optical calculation component of GCMs.

Second, we have already conducted preliminary sensitivity experiments with the WRF model over the East Asian region to test the applicability of FACM at a larger spatial scale. The results show that after accounting for aerosol fractal morphology, the model produces non-negligible effects on aerosol optical depth, surface shortwave radiation, and surface temperature over East Asia, indicating that FACM has clear potential for improving aerosol radiative forcing estimates. However, due to the scope and workload of this paper, these results are not formally presented in the main text but are instead included in the Supplementary Material S2 (Figures S3–S7).

We greatly appreciate the reviewer's professional and forward-looking suggestion, and we hope that in the future we or other researchers can, based on the model tool provided in this paper, further systematically evaluate the applicability of FACM in GCMs, including different D_f parameterization schemes, coupling with aerosol mixing-state schemes, and impacts on global radiative forcing estimates.

Corresponding revision plan:

In the last paragraph of the **Conclusions**, add a brief discussion on the transferability of FACM and its prospects for application in GCMs, with a reference to Supplementary Material S2:

"Furthermore, FACM has good transferability. The scheme relies on few parameters and is physically well-defined, making it readily embeddable into the aerosol optical modules of other regional models or global climate models. We have conducted preliminary sensitivity experiments over East Asia, and the results show that fractal aerosols have non-negligible impacts on aerosol optical depth, surface shortwave radiation, and surface temperature, demonstrating the potential of FACM for improving aerosol radiative forcing estimates. Due to the scope and workload of this paper, these

results are not presented in the main text; see Supplementary Material S2 for details. In future work, we will systematically evaluate the applicability of FACM in GCMs, including different fractal dimension parameterization schemes and their coupling with aerosol mixing-state schemes."

Reference in this response to comments:

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