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Comment on "Could We Achieve the On-Line Measurements of the Optical Fractal Dimensions of Black Carbon?" by G. Zhao, M. Hu, W. Lin, Y. Kuang, J. Sun, L. Zeng, and C. Zhao

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Key Points:

- Optical scattering measurements of the fractal dimension and size of black carbon have been demonstrated
- Such optical measurements could be used to develop a low-cost online instrument

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Abstract Zhao et al. (2025, https://doi.org/10.1029/2024GL112332) state the need for measuring the fractal dimension of black carbon aggregates online and proceed to propose and demonstrate a novel method to derive this fractal dimension from measured mobility parameters and mass concentrations. Their summary of the previous state-of-the-art is that the fractal dimension could not be measured online. Here, we briefly review the literature on the current state-of-the-art for light scattering experiments and clearly establish that optical laboratory measurements of aggregate fractal dimension and size have been performed. Furthermore, we agree with Zhao et al. (2025, https://doi.org/10.1029/2024GL112332) on the importance of developing and utilizing online instruments for such measurements and argue that optical scattering measurements can do so at low cost.

Plain Language Summary Some ambient aerosols, such as black carbon agglomerates have complex and evolving fractal-like structures that greatly affect their radiative forcing and influence on climate change. Therefore, there is a need for online monitoring of their structure as pointed out by Zhao et al. (2025, https://doi.org/10.1029/2024GL112332), who have also proposed and demonstrated a method for this, arguing the novelty of their method. Here, we demonstrate that existing optical light-scattering experiments have already made such measurements and are well suited for the development of low-cost optical online monitors.

1. Introduction

Zhao et al. (2025) emphasize the need for being able to measure the fractal dimension (D_f) of black carbon (BC) aggregates online. This would be very useful for studying the transformation of BC morphology and optical properties during atmospheric aging. However, they state multiple times that "*D_f cannot be measured online*" and that "*D_f is always measured by using the electron microscope, which means that the D_f is not able to be measured online*" and continue to propose and demonstrate a novel method to derive D_f from measured BC mobility parameters and mass concentrations. We strongly disagree with the statement that previously BC's fractal dimension D_f could not be measured online.

Their work (Zhao et al., 2025) articulates well the need for online characterization of BC physical properties such the fractal dimension and size. Here, we cite and briefly discuss a great deal of previous, peer-reviewed literature regarding optical scattering and mobility properties of fractal aggregates in general and BC in particular. This literature lays the foundation for viable online measurements of D_f and illustrates experimental procedures and results.

2. Optical In Situ Measurements of the Fractal Dimension of Fractal Aggregates

There is a long history of publications demonstrating that the Rayleigh-Debye-Gans (RDG) approximation for fractal aggregate light scattering and absorption works very well (Cai et al., 1993; Köylü & Faeth, 1994a, 1994b; Sorensen, 2001; Sorensen et al., 2018; Wang & Sorensen, 2002). Much of this work studied BC aggregates. In particular, Sorensen (2001) reviewed work up to 2001 and put the RDG theory for fractal aggregates (RDGFA) on a solid foundation. The theory was tested with experiments by Wang and Sorensen (2002) and theoretically by

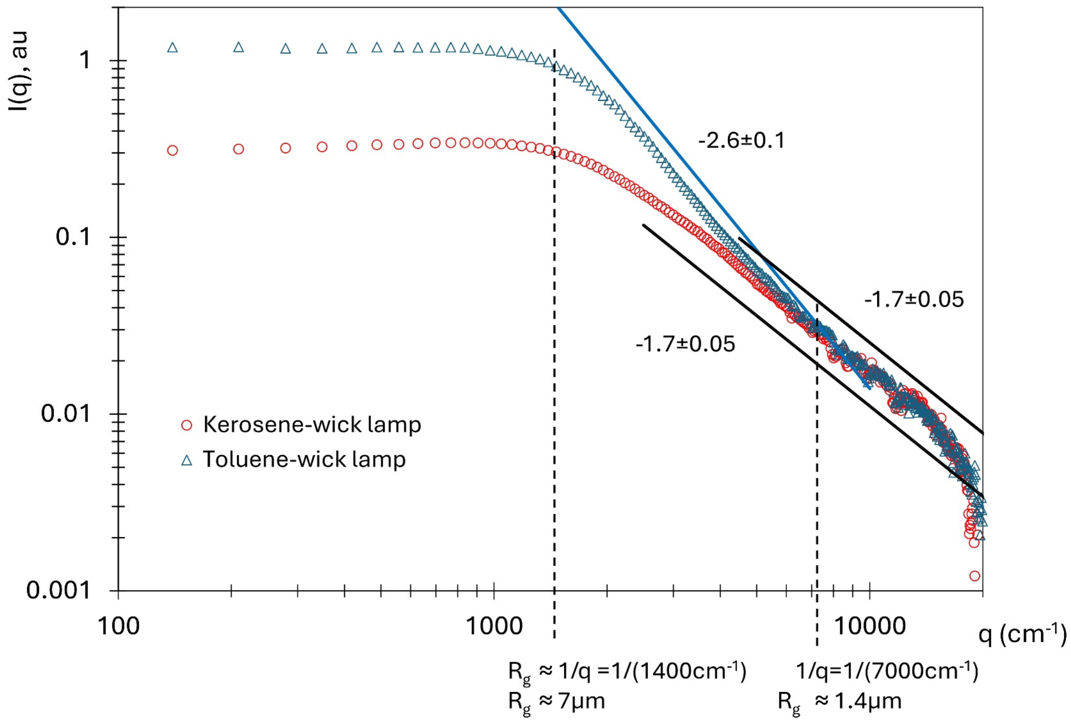

Figure 1. Optical structure factors, $I(q)$ versus q, for soot aerosols emanating from a wick lamp. The aerosols were produced by burning two different fuels, kerosene and toluene. Both structure factors show a constant forward scattering lobe at small q. Then, with increasing q, a curving Guinier regime follow by power law regimes. Both Guinier regimes are near $q \approx 1{,}400\ \mathrm{cm}^{-1}$, the inverse of which is 7 microns. This is the size of the soot particles. The slopes of the power laws are−1.7 to imply the soot particles are fractal aggregates with fractal dimensions of 1.7. The toluene flame shows two slopes. The−2.6 slope implies that the soot particles are superaggregates with a fractal dimension of 1.7 at length scales less than $q^{-1} = 1.4$ microns, and a fractal dimension of 2.6 at length scales between 1.4 and 7 microns.

Sorensen et al. (2018), finding the RDGFA theory accurate to 10%–20%. Most recently it has been summarized by Sorensen (2022).

With RDG in mind, measurements of scattered intensity over a broad range of angles, including those approaching zero as closely as possible, yields both the fractal dimension and the mean radius of gyration of the scattering entities. To do this, the scattered intensity data are plotted in Q-space (Sorensen, 2022). Q-space analysis involves converting the scattering angle θ to the magnitude of the wave vector q, via

$$q = (4\pi/\lambda)\sin(\theta/2), \tag{1}$$

where λ is the wavelength of the scattered light. Note that the dimension of q is inverse length, and q depends on the easily controlled scattering angle θ during the experiment. By altering the scattering angle, the light examines various length scales of the scattering particles, allowing for the measurement of their structure and size (Sorensen, 2001).

With q calculated, the next step in Q-space analysis involves plotting the scattered intensity $I(q)$ versus q in a log-log fashion. Thereby, one finds a regime at small q of constant scattering, the forward-scattering lobe. With increasing q, $I(q)$ will decrease as it enters the Guinier regime. Analysis of this Guinier regime yields the mean particle radius of gyration. Finally, if the particles are fractal aggregates, at even larger q, $I(q) \sim q^{-D_f}$, and a power law will appear, linear with a slope of $-D_f$ on the log-log plot. This simple procedure has appeared in many publications in the past ≈40 years. Given its simplicity, it is entirely amenable to on-line applications. An example of Q-space analysis of light scattered from an in-flame soot aerosol is given in Figure 1.

3. Relationship Between the Mobility Parameter (D_m) and the Radius of Gyration (R_g)

There is also an extensive literature regarding the mobility of fractal aggregates. On this topic, Sorensen (2011) gives us another extensive review of the experimental and theoretical literature covering three decades prior to 2011. Aggregates formed via either diffusion and reaction limited cluster-cluster aggregation processes (DLCA and RLCA, respectively) were considered. These processes form aggregates with fractal dimensions and prefactors of ≈1.78 and ≈1.3 (DCLA) and ≈2.1 and ≈0.94 (RCLA), respectively. Moreover, the entire Knudsen number range from continuum to free molecular was reviewed. The review provides a straightforward summary of the mobility radius contained in Sorensen (2011), Figures 5 and 6.

Zhao et al. (2025) presented theoretical results for the mobility radius of fractal aggregates with dimensions ranging as $2.8 \gtrsim D_f \gtrsim 1.4$. This is a broader range than previously considered by Sorensen (2011), but both results are consistent in the range where they overlap.

4. Online Measurements

Online measurement devices for fractal dimension and size can be built, as demonstrated by Heinson et al. (2019). We believe that given the state-of-the-art as described above, now is a propitious time to develop online optical field instruments. Moreover, we recommend as a guide the work of Gautam and Sorensen (2021). Although this work presents a laboratory setup, it demonstrates the importance of small scattering angles, as small as 0.32^0, to measure particles with diameters of up to ~30 μm. It also includes large angles out to 178^0 where glory-like scattering occurs, and advocates coupling Q-space analysis with linear plotting versus the scattering angle to get a complete vision of the scattering. Notice that optical scattering measurements are generally capable of performing nondestructive online measurements at low cost.

5. Conclusions

We have reviewed the current state-of-the-art for light scattering experiments for any particle shape. We have emphasized scattering by fractal aggregates, such as BC. Our review emphasizes that online scattering measurements of the fractal dimension and size of BC aggregates are very possible. We also found agreement of the theoretical calculations of Zhao et al. (2025) for fractal aggregate mobility with a published review of empirical data (Sorensen, 2011).

Conflict of Interest

The authors declare no conflicts of interest relevant to this study.

Data Availability Statement

Data were not used, nor created for this work.

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References

Cai, J., Lu, N., & Sorensen, C. M. (1993). Comparison of size and morphology of soot aggregates as determined by light-scattering and electron microscope analysis. *Langmuir*, *9*(11), 2861–2867. https://doi.org/10.1021/la00035a023

Gautam, P., & Sorensen, C. M. (2021). A wide range (0.32°–177.6°), multi-angle light scattering setup and concomitant analysis method. *Review of Scientific Instruments*, *92*(11), 113105. https://doi.org/10.1063/5.0068318

Heinson, Y. W., Sorensen, C. M., & Chakrabarty, R. K. (2019). Portable device to determine particle asymmetry parameter. *Optics Express*, *27*(7), 10152–10158. https://doi.org/10.1364/OE.27.010152

Köylü, Ü. Ö., & Faeth, G. M. (1994a). Optical properties of overfire soot in buoyant turbulent diffusion flames at long residence times. *Journal of Heat Transfer*, *116*(1), 152–159. https://doi.org/10.1115/1.2910849

Köylü, Ü. Ö., & Faeth, G. M. (1994b). Optical properties of soot in Buoyant laminar diffusion flames. *Journal of Heat Transfer*, *116*(4), 971–979. https://doi.org/10.1115/1.2911473

Sorensen, C. M. (2001). Light scattering by fractal aggregates: A review. *Aerosol Science and Technology*, *35*(2), 648–687. https://doi.org/10.1080/02786820117868

Sorensen, C. M. (2011). The mobility of fractal aggregates: A review. *Aerosol Science and Technology*, *45*(7), 765–779. https://doi.org/10.1080/02786826.2011.560909

Sorensen, C. M. (2022). *Light scattering and absorption by particles: The Q-space approach*. IOP Publishing. https://doi.org/10.1088/978-0-7503-5157-7

Sorensen, C. M., Yon, J., Liu, F., Maughan, J., Heinson, W. R., & Berg, M. J. (2018). Light scattering and absorption by fractal aggregates including soot. *Journal of Quantitative Spectroscopy and Radiative Transfer*, *217*, 459–473. https://doi.org/10.1016/j.jqsrt.2018.05.016

Wang, G., & Sorensen, C. M. (2002). Experimental test of the Rayleigh-Debye-Gans theory for light scattering by fractal aggregates. *Applied Optics*, *41*(22), 4645–4651. https://doi.org/10.1364/AO.41.004645

Zhao, G., Hu, M., Lin, W., Kuang, Y., Sun, J., Zeng, L., & Zhao, C. (2025). Could we achieve the on-line measurements of the optical fractal dimensions of black carbon? *Geophysical Research Letters*, *52*(4), e2024GL112332. https://doi.org/10.1029/2024GL112332