Comment 1: Romshoo et al. (egusphere-2023-2400) simulated bare soot aggregates by diffusion-limited cluster aggregation (DLCA) and coated soot by adding spherical coatings around the spherical monomers of the DLCA aggregates. The authors then applied machine-learning models to interpolate numerically accurate MSTM calculations of the aggregates' optical properties.

The approach taken by the present authors was taken by two different studies previously. Luo et al. (2018) considered soot aggregates but not coatings. Lamb and Gentine (2021) considered uncoated and coated soot, using a coating model almost identical to the present authors. The authors erroneously wrote that Lamb and Gentine do not consider coating", but the only difference in their approach is an insignificant change in the machine-learning model. For this reason, I have to recommend rejection of the present manuscript.

Response: Thank you for your comment. According to our understanding, Lamb and Gentine (2021) generated the optical properties of bare BC fractal aggregates using a graph neural network (GNN). We found strong evidence that they did take coating into account. On page 2 of the paper is written:

"Here, we show the optical properties of bare BC with complex morphology can be accurately predicted with a graph neural network (GNN) by representing BC fractal aggregates as networks of interacting spheres. GNNs are recently developed machine learning algorithms that learn on graph-structured data sets, allowing models to include arbitrary relational information directly."

Furthermore, on Page 8, the authors wrote, "As a proof of concept, we have trained a GNN to predict the optical properties of bare BC fractal aggregates with a range of different fractal parameters."

On page 8 of the paper, **as an outlook**, it is written that "The GNN approach provides an obvious extension to internally mixed aerosols (Fig. 1), as the thickness of coatings and their indices of refraction or organic fraction could be included as additional node-level features (in the thinly coated case) or graph-level features (for the thickly coated case)."

The above sentence could have led to an easy misinterpretation, but we checked that there is no indication of coating in Table S1 of the parameters provided in the supplementary material. They also used refractive indices with an imaginary part higher than 0.4, which is typical for bare BC aggregates. The authors of Lamb and Gentine (2021) mean that others can extend their approach to include other node features like coating. Therefore, we respectfully disagree with your comment.

Please find the recently published version of Lamb and Gentine (2021) below:

https://www.nature.com/articles/s41598-023-45235-8

Comment 2: Independently, I also cannot recommend publication of this manuscript as the coating model is completely unrealistic. No experiment has ever observed coated soot to retain its original shape while adding spherical coatings to the monomers. Romshoo et al., and several others have already published this model, but it contradicts dozens of smog chamber and field studies using electron microscopy, which all observed restructuring. There is no value in using machine-learning algorithms to interpolate the results of an inaccurate model.

Response: Thank you for your comment. Atmospheric soot undergoes various processes, including a possible restructuring after emission, depending on multiple factors such as geographical location, atmospheric chemistry, and meteorology (Fig. 1 adapted from Sedlacek et al., 2022). The lifecycle of soot particles is not captured properly by global climate models. Current state of art for representing atmospheric soot particles focusses on spherical aged particles. At any point in the atmosphere, BC can exhibit a wide range of morphologies showing diversity at different locations (Fig. 2 taken from Fu et al., 2012). It was observed that aged transported soot can retain its fractal morphology 500 to 1000 km downwind of emission sources (Fig. 3 adapted from Sun et al., 2020). The model provided in this study was designed to simulate the optical properties for the entire BC lifecycle capturing the transition between fresh fractal to aged spherical particles.

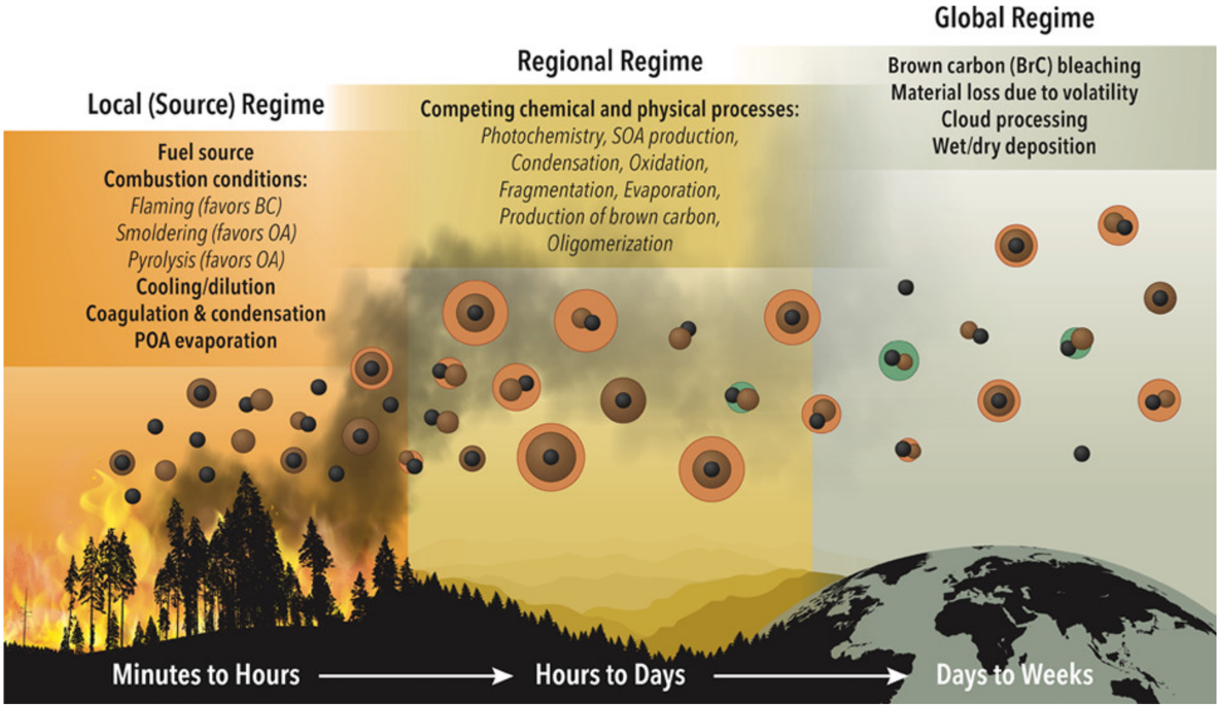

Figure 1. Lifecycle of BC particles adapted from Sedlacek et al., 2022.

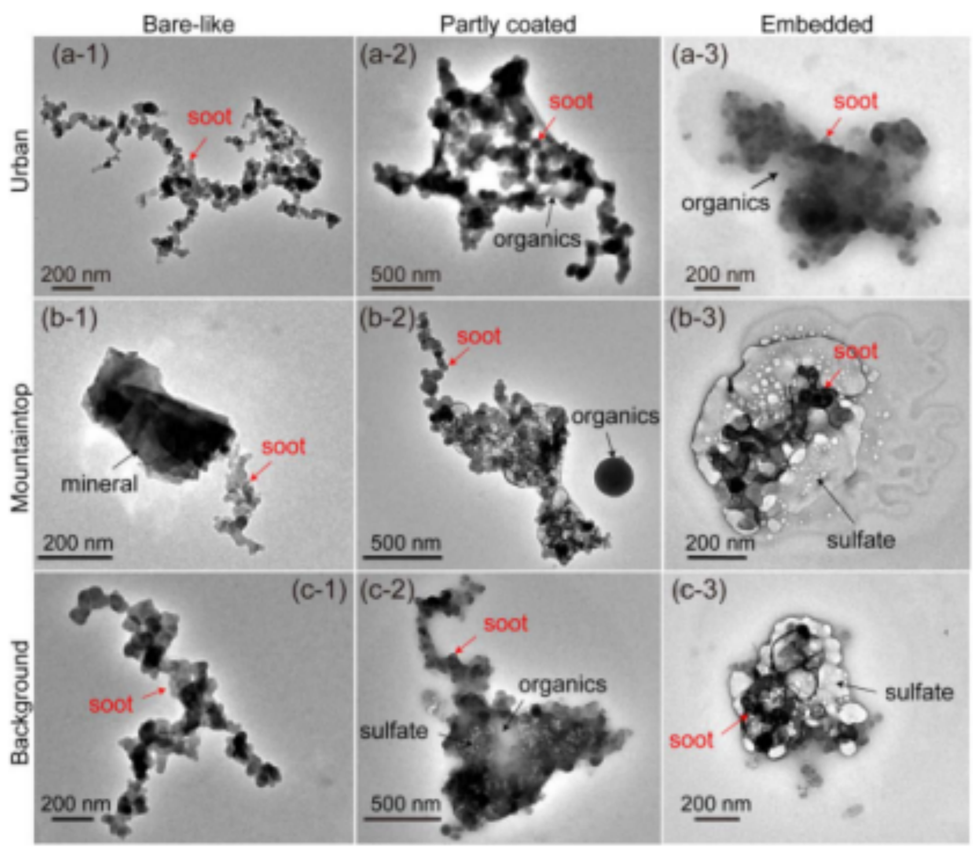

Figure 2. Transmission electron microscopy (TEM) images adapted from Fu et al., 2012 showing wide range of BC morphologies at different locations.

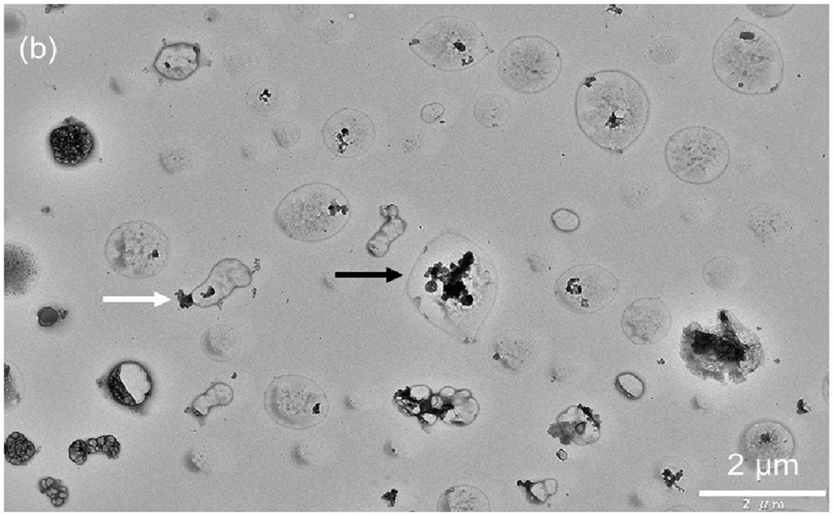

Figure 3. Results from TEM analysis showing that aged transported soot can retain its fractal morphology 500 to 1000 km downwind of emission source, taken from Sun et al., 2022.

The restructured soot is most representative of the “embedded” soot. However, BC particles can have thin, medium, or partial coatings as well. For this reason, we provide a ML algorithm that provides optical properties for all possible ranges of fractal dimensions between 1.5 and 2.9. Furthermore, it depends on the user to choose a suitable fractal dimension and fraction of coating to represent the BC particles they want to simulate. For example, for restructured aged particles, the user can choose a fractal dimension higher than 2.5, and coating fractions higher than 50 %.

The coating model used in this study is called the "closed-cell model," the results showed good comparability with the realistic coating model (Kahnert 2017). It was necessary to use this coating model due to the limitations of the MSTM code that was used for generating the optical properties. We agree that a more sophisticated coating model would be a good choice, but it requires more complex scattering models, such as Discrete Dipole Approximation (DDA), which is computationally expensive. With the DDA method, generating elaborate datasets for training ML algorithms is not feasible.

Simulations of BC's optical properties are required for global climate models. Presently, the simplistic Mie core-shell model is used for BC particles, representing the aged portion of the BC lifecycle. Although a more realistic coating model would be ideal, our ML method offers a robust solution for predicting the present scenario due to the above-discussed limitations. We provide a method that predicts the optical properties of a wide range of ambient soot particles with high accuracy. Using this method, we can overcome the limitations of the simplistic core-shell model, which only represents aged BC particles. Furthermore, calibration of light absorption measurement devices mostly is done with fresh soot. We can make the link to atmospheric relevant absorption by simulating mass absorption cross-sections and light absorption enhancement factors.

Therefore, the results of this study are valuable for the simulation of realistic scenarios, despite the model limitations. We acknowledge that there is scope for future studies to extend such an ML-based approach using other morphological models of BC and coating positions. Reviewer’s feedback will be carefully considered in the revised manuscript, providing a detailed explanation of our approach and its limitations in the revised manuscript.

References:

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