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AS&T Article Highlight: Mechanisms of Soot-Aggregate Restructuring and Compaction

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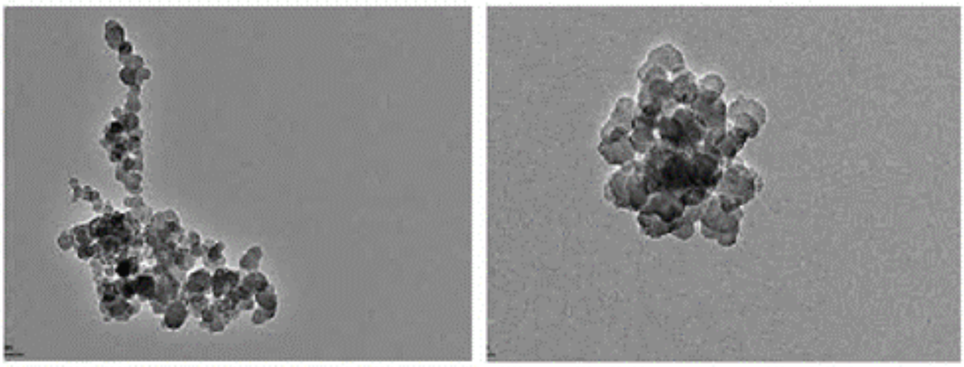
Soot aggregates are delicate – in the atmosphere, contact with a condensing liquid causes their elaborate tiny fractals to collapse, compacted by capillary tension. The scattering and absorption of shortwave radiation by black carbon aerosols change as the fresh soot is coated and collapses into denser agglomerates. The mechanism of soot collapse is important when considering how light absorption changes over time in an aging air parcel. Does soot collapse via nonuniform vapor deposition and pooling in the aggregate's crevices? Or does the soot collapse during evaporation of a fully enveloping liquid layer?

Corbin et al. have performed careful experiments to address all possible mechanisms of soot collapse, and provide, alongside their measurements and analysis, a very good review of the history of the topic and discussion of prior work (which has generally concluded either that soot collapses upon condensation or that soot collapses during evaporation). They show that aggregates collapse with condensation, which begins at active sites in the crevices and then spreads around the aggregated spherules, pulling them together via line tension at the three-phase interface (the line where solid soot, air, and the liquid meet). How, then, do so many studies see evidence of fresh fractal aggregates in liquid that collapse upon droplet evaporation? The present study explores this apparent dichotomy at length. A high contact angle condensate (namely water) is not efficiently deposited on soot surfaces. As the gas-phase saturation rises above a critical point, the nearly dry soot aggregate undergoes droplet nucleation, preventing it from experiencing prolonged capillary tension at its joints. Thus, the fractal particle, which at the necks of the sintering spherules is covalently bonded, retains its lacy form while immersed. It may then collapse during the evaporation of the droplet. This level of water supersaturation does not usually occur in the atmosphere.

Furthermore, the study examine how soot aggregates might retain their fluffy form. If the condensing liquid is highly viscous, essentially freezing onto the soot surfaces, the tension remains but a kinetic limitation inhibits both flow and soot collapse. Evaporation puts increased tension on any soot particles – whether compacted by the condensation process or preserved in a fractal shape. The evaporative restructuring is impactful for soot that has escaped compaction during wetting. The only mechanism for fresh soot to take up a condensate, lose it again to the vapor phase, and still retain its unfolded form, is to take up a highly viscous liquid that subsequently sublimates.

All of these mechanisms were demonstrated by the authors using a suite of online instruments and offline analyses. The experiments, along with the succinct review of over 150 prior contributions, merit the attention of researchers addressing soot in any context.



[Source: Corbin et al., 2023, Figure S4, https://doi.org/10.1080/02786826.2022.2137385]

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