The superaggregate form of soot from wildfires and its impact on direct forcing

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Atmospheric soot has been implicated in rapid global warming, accelerated melting of glaciers, changing monsoon patterns, and degradation of human health and the environment. On a regional-to-global scale, wildfires emit up to 63% of total soot mass into the atmosphere, contributing to greater than threefold uncertainty in current estimates of climate forcing. This large uncertainty is due to poor understanding of wildfire-emitted soot microphysical properties and their parameterizations in models and satellite retrieval algorithms. For several decades, researchers have made efforts to characterize these properties for soot emitted from small-scale, controlled combustion systems—such as prescribed and slash burns—as a function of various process parameters such as fuel type, temperature and scale of flaming phase, environmental conditions driving the flame, and interrelationships of these parameters. Consequently, the current view holds that soot is formed via the cluster-dilute aggregation mechanism in wildfires and emitted as sub-micrometer size aggregates of fractal dimension $D_f \approx 1.8$. Here we report ubiquitous presence of soot superaggregates (SAs) in the outflow from two wildfires in India and California. SAs are aggregates of cluster-dilute aggregates with characteristic $D_f \approx 2.6$ and $D_m > 1\mu m$ that form via the cluster-dense aggregation mechanism. We present additional observations of soot SAs in wildfire smoke-laden air masses over New Mexico and Mexico City. Using numerically-exact *T*-Matrix optical modeling, we estimate soot SAs to contribute, per unit optical depth, up to 35% less atmospheric warming than freshly-emitted aggregates, about 25% more warming than aged aggregates, and ≈90% more warming than spherical particles currently used in climate models. Our findings imply significant and previously unaccounted-for impacts by soot SAs from wildfires on climate forcing as well as potentially deleterious effects on human health and the environment.