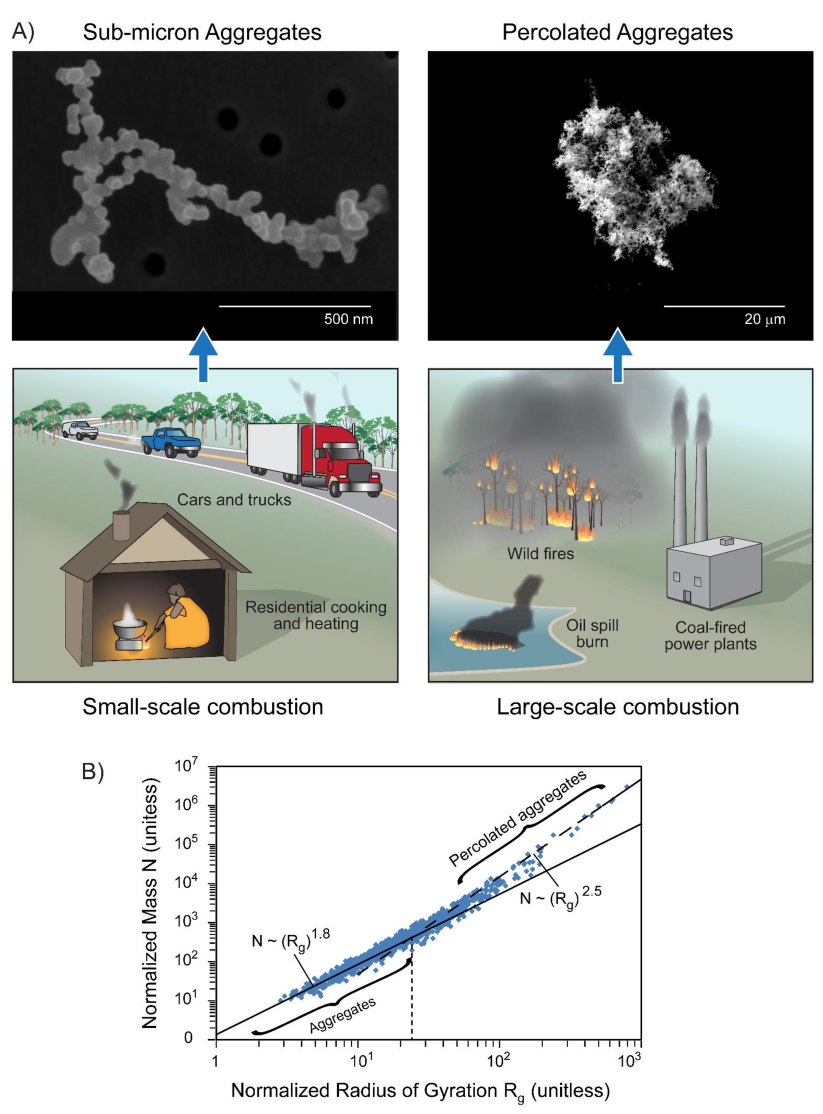
Radiative Properties of Soot Superaggregates in the Infrared

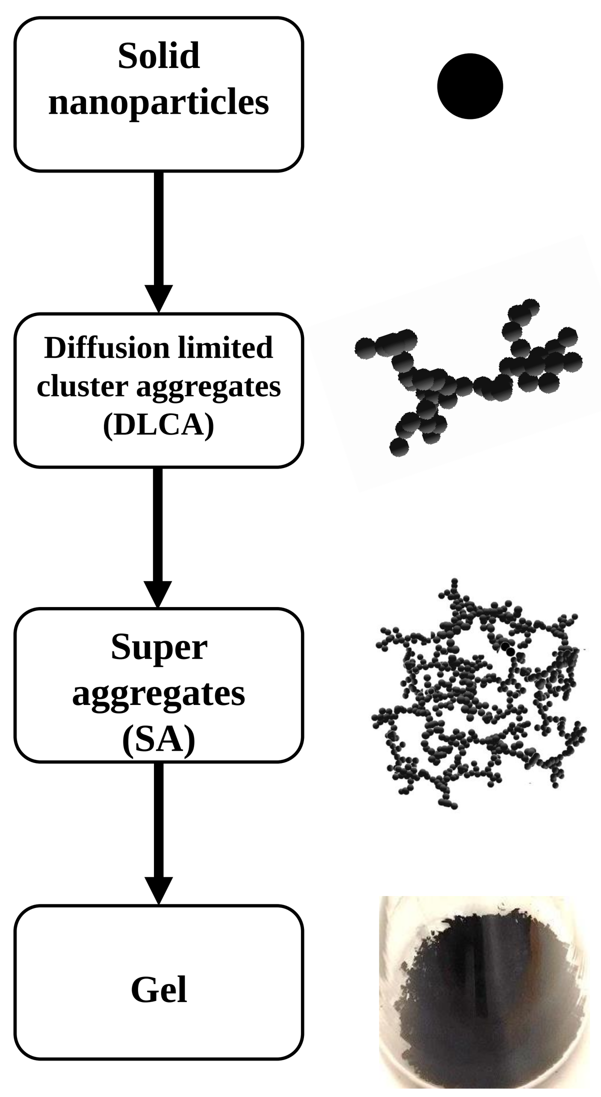
Yang Yu, Ian J. Arnold and Rajan K. Chakrabarty*
Aerosol Impact & Research (AIR) Laboratory
*PI Contact Address: chakrabarty@wustl.edu

Soot is one of the largest man-made contributors to global warming and its influence on climate has been greatly underestimated. This underestimation is due to poor understanding of the microphysical properties of soot and its parameterization in climate models and satellite retrieval algorithms. Very recently, researchers observed a new kind of soot particle "super aggregates", emitted from large-scale forest fires globally. The morphology of these particles is described by a mass fractal dimension ≈ 2.6, mobility diameter > 1 μm, and aerodynamic diameter in the range of 0.5 - 20 μm. Given their large size, it is expected that these soot superaggregates would impact radiative forcing in the longer wavelengths (i.e. infrared (IR)) of the incoming solar spectrum. Here, we quantitatively investigate the microphysical and optical properties of soot superaggregates using controlled laboratory-scale experiments. Soot superaggregates were produced in our laboratory using a novel diffusion flame aerosol reactor operated in a negative gravity. Next, an Integrated Photoacoustic-Nephelometer (IPN) System using a 1047nm laser source was built to simultaneously measure absorption and scattering coefficient of these particles in real-time. With the parameters above, Mass Absorption Cross Section (MAC), Mass Scattering Cross Section (MSC), Mass Extinction Cross Section (MEC) and Single Scattering Albedo (SSA) could be calculated. Our results show that soot superaggregates have distinct physical and optical properties compared to conventional sub-micron soot aggregates, and could significantly impact direct radiative forcing in the atmosphere.

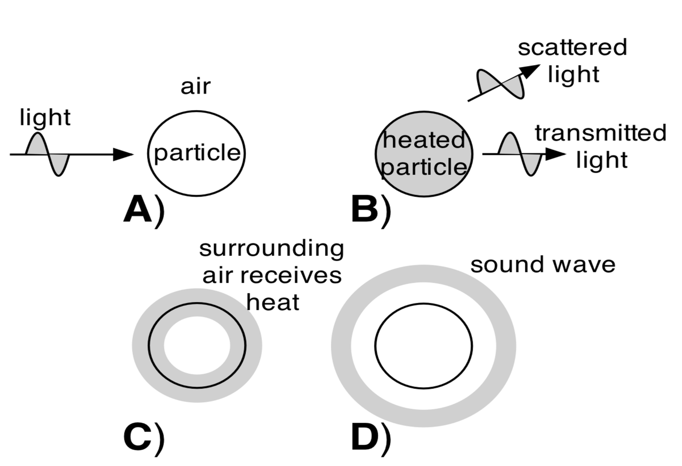
Background

- **Different Morphologies of Atmospheric Soot**



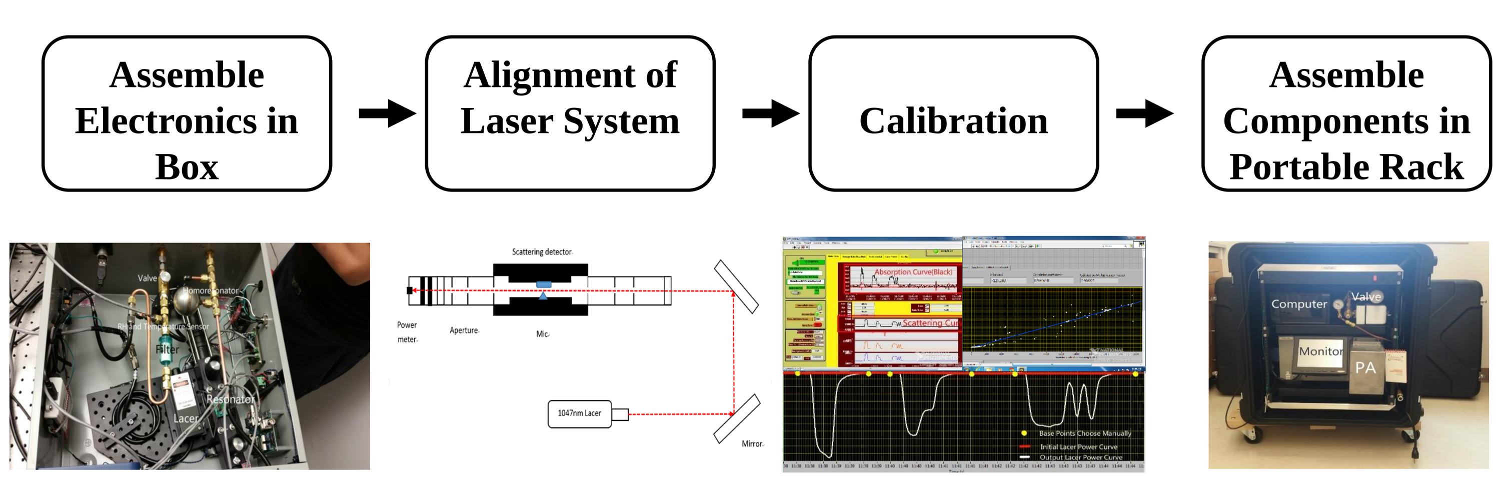


- **Photoacoustic Effect**



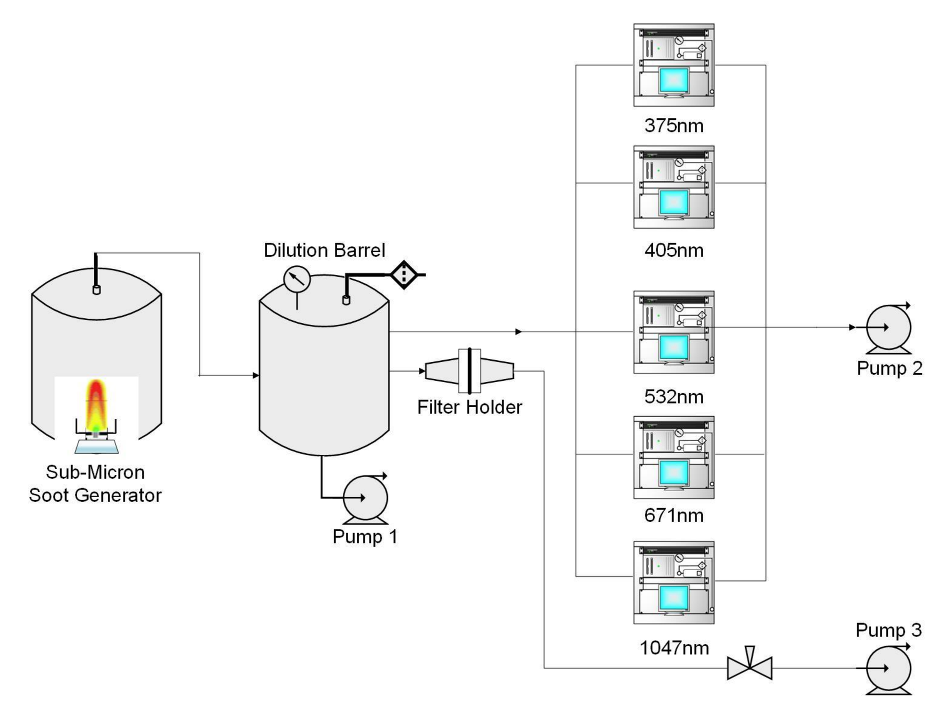
When electromagnetic wave interacts with a particle, part of the light will be scattered and part will be absorbed by particle. Light absorbed by particle results in temperature increase of particle. The temperature change $\Delta T(t)$ of a particle of radius (r) illuminated by a laser beam of irradiance I is given by $\Delta T(t) = T_\infty[1-\exp(-t/\tau)]$ and the eventual temperature (T_∞) is $T_\infty = \Delta T(t=\infty) = \frac{I Q_{abs} r}{4\kappa_a}$. Heat transfers by conduction from the particle to the surrounding air, expanding the surrounding air and creating a pressure distribution or sound wave. At the same time, the sound wave signals will be recorded by microphone and the scattering signal will be recorded by cosine-weighted scattering detector.

- **Building 1047nm Photoacoustic-Nephelometer System**



Experimental

- **Sub-Micron Soot Set Up**



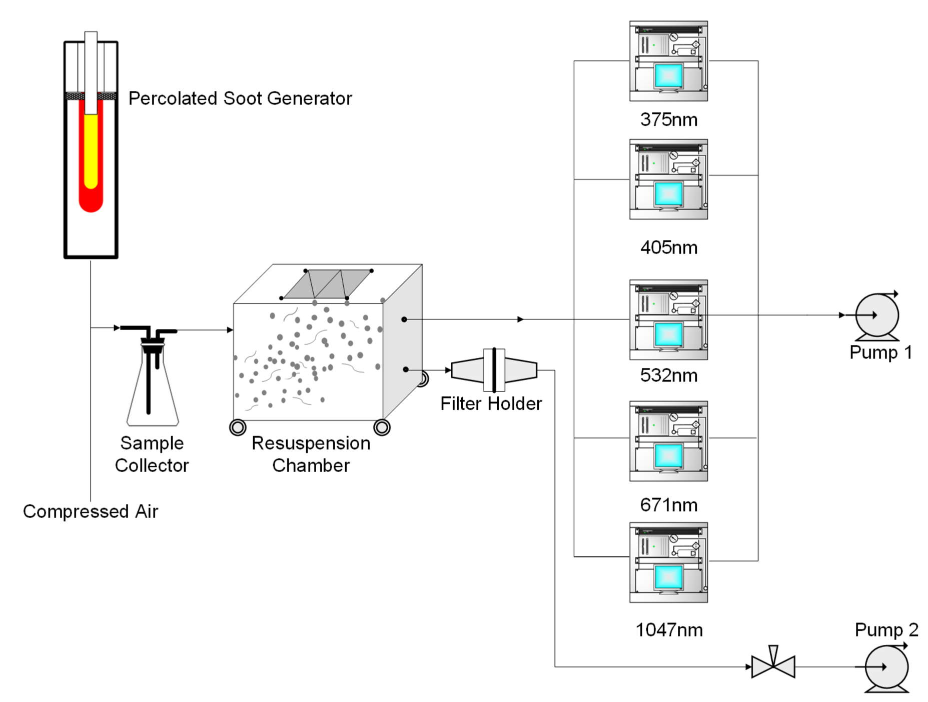
Set-up includes a soot generation system, a dilution system, a collection system and a measurement system.

A Dilution Barrel was used to control the concentration of soot and decrease the effect of NO_x.

To measure the mass flow rate of Sub-Micron soot, a 47mm in-line filter holder with PTFE membrane was integrated with the dilution barrel.

optical properties were measured by a series of PA Systems (375nm, 405nm, 532nm, 671nm and 1047nm).

- **Super Aggregate Soot Set Up**



Set-up includes a soot generation system, a resuspension system, a collecting system and a measurement system. The collecting system and measuring system are same as these in Sub-Micron Soot set-up.

Resuspension system consists of a flask and a resuspension chamber. Pulverized percolated aggregate soot was placed into the flask and shoot into the resuspension chamber by compressed air.

- **Size Distribution of Super Aggregate Soot**

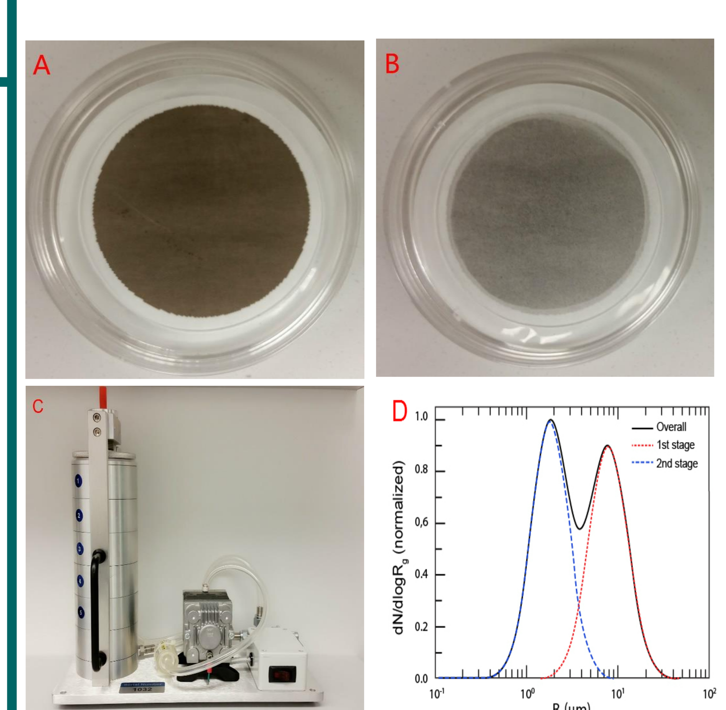


Figure (A): Image of Telfon filter sample collected from Sub-Micron Soot

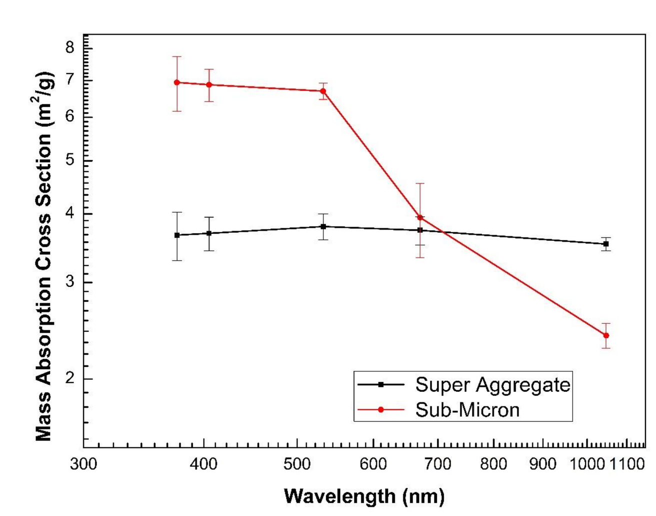
Figure (B): Image of Telfon filter sample collected from Super Aggregate Soot

Figure (C): Image of Microanalysis Particle Sampler Model MPS-6

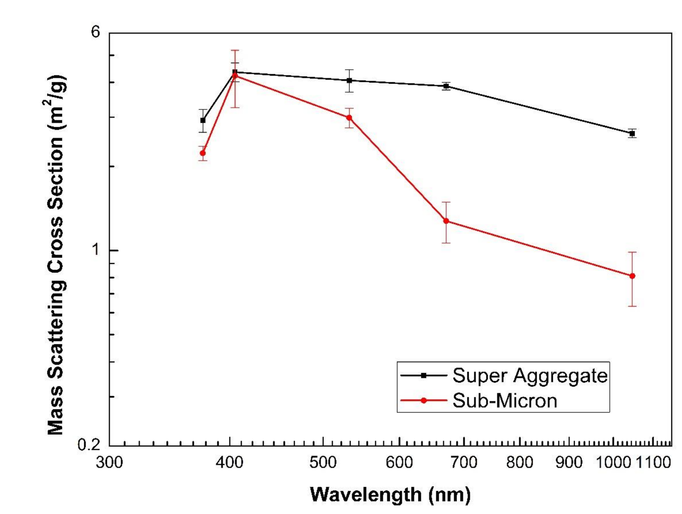
Figure (D): Size Distribution of Super Aggregate Soot

Results

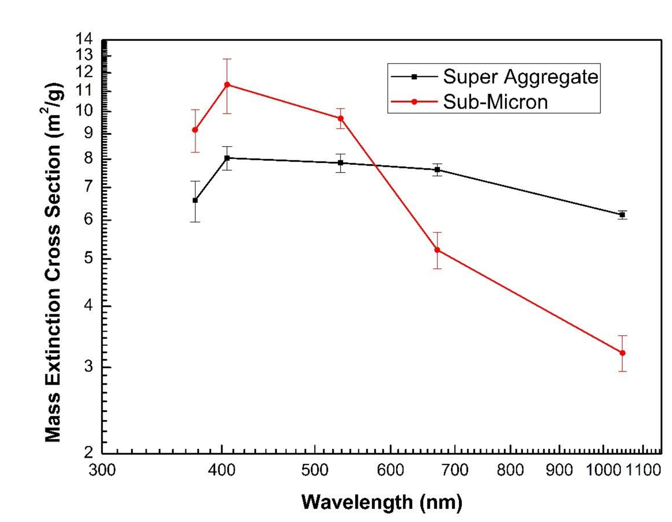
UV-Vis-NIR Optical Properties



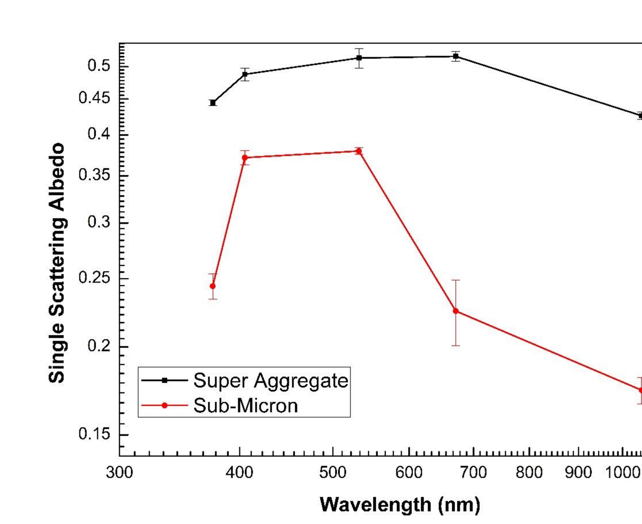
$$MAC\left(\frac{m^2}{g}\right) = \beta_{abs}(m^{-1}) * \frac{f_V(m^3/s)}{f_m(g/s)}$$



$$MSC\left(\frac{m^2}{g}\right) = \beta_{sca}(m^{-1}) * \frac{f_V(m^3/s)}{f_m(g/s)}$$



$$MEC\left(\frac{m^2}{g}\right) = \beta_{ext}(m^{-1}) * \frac{f_V(m^3/s)}{f_m(g/s)}$$



$$SSA = \frac{\beta_{sca}(m^{-1})}{\beta_{sca}(m^{-1}) + \beta_{abs}(m^{-1})}$$

Absorption Angstrom Exponent

Sub-Micron Aggregate				
λ(nm)	375-405	405-532	532-671	671-1047
MAC	0.3438	0.231	2.285	1.113
MSC	-8.2921	1.267	3.664	1.019
MEC	-2.7671	0.583	2.266	1.089

Percolated Aggregate				
λ(nm)	375-405	405-532	532-671	671-1047
MAC	0.1027		0.067	0.131
MSC	-5.1679	0.231		0.873
MEC	-2.5978	0.108		0.478

Infrared

Reference: Chakrabarty, Rajan K., et al. "Soot superaggregates from flaming wildfires and their direct radiative forcing." Scientific reports 4 (2014).