Group Seminar:

Morphological Properties of Atmospheric Aerosol Aggregates

University of Connecticut

Focus of the Study

- Importance of morphology of atmospheric aggregates in public health, water vapor nucleation and absorption and scattering of light.
- To study and collect ultrafine particles (smaller than 0.1 μm) in urban and rural areas.
- To obtain the mass-based fractal dimension (D_f) and prefactor (A) for aggregates.
- To obtain better estimates of atmospheric aggregate residence times, transport, and deposition in lung, optical extinction, and heterogeneous nucleation.

Experimental Methods

- Sampling Sites: UCLA campus, San Jacinto, CA, Santa Monica, CA, UCLA neighborhood, and Reseach Triagle Park, NC.
- Transmission Electron Microscope (TEM) grids fitted in the last two stages of a single-jet eight–stage low pressure impactor (LPI) for a period of a few minutes. Stages have a 50% aerodynamic diameter cutoffs of 4.0, 2.0,1.0, 0.5, 0.26, 0.12, 0.075, and 0.05 μm (1 l/min).
- Differential Mobility Analyzer (DMA) and Condensation Particle Counter (CPC) system (TSI, Models 3070 and 3010).

Mass-Based Fractal Dimension

$$N_p = A\left(\frac{R_g}{R_o}\right)^{D_f}, \qquad [1]$$

- D_f is the fractal dimension, N_p is the number of primary particles in the aggregate, A is a dimensionless prefactor, R_o is the primary particle radius, and R_g is the characteristic radius of the aggregate, taken to be the radius of gyration.

$$\log N_p = \log A + D_f \log R_g - D_f \log R_o \qquad [2]$$

- Where $R_g=[(1/M)\Sigma(m_i r_i)]^{1/2}$, m_i is the mass of the ith primary particle, M is the total aggregate mass $=\Sigma m_i$ and r_i is the distance of the ith primary particle from the center of mass.
- Fractal-like aggregates with $1.35< D_f <1.89$. Possibly non-fractal particles with $D_f >2$.

D_f for Soot Aggregates and Size Collection Ranges

Table 1. Average values of D_f for soot aggregates from laboratory sources

Source	D_f	Method of analysis	Reference
Acetylene flame	1.5–1.6	Electron microscopy	7
Butane flame	1.87–2.19	Light scattering	10
Diesel engine	2.1–2.9	Mobility analyzer	11
Spark ignition engine	2.2–3.0	Mobility analyzer	11

Table 2. Comparison of aggregate size ranges, nm, for LPI and DMA-CPC

LPI	DMA-CPC
50–75 (stage 8)	51–71
75–120 (stage 7)	71–100

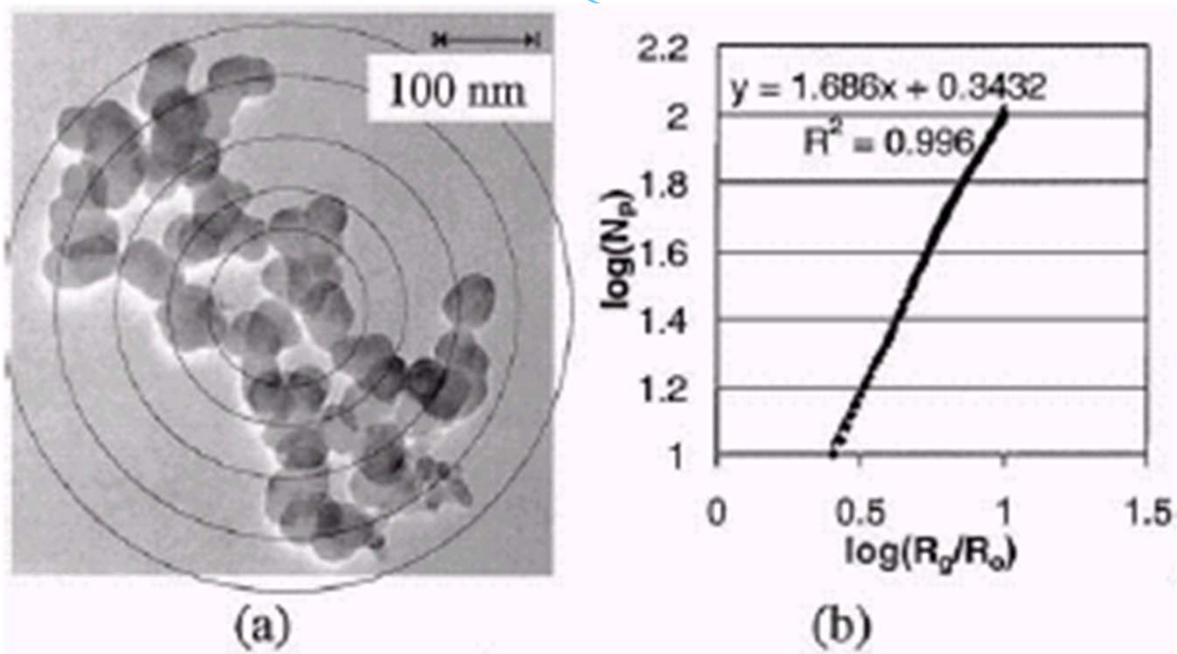

Fig. 1. Fractal analysis of an aggregate for stage 7. (*a*) An electron micrograph of an aggregate imported into COREL DRAW. The concentric circles correspond to different r_i. (*b*) The fractal dimension (D_f) was determined from the slope of the log–log plot of the normalized radius of gyration and number of primary particles. The normalizing radius R_o is based on a volume average, and *A* is determined from the *Y* intercept.

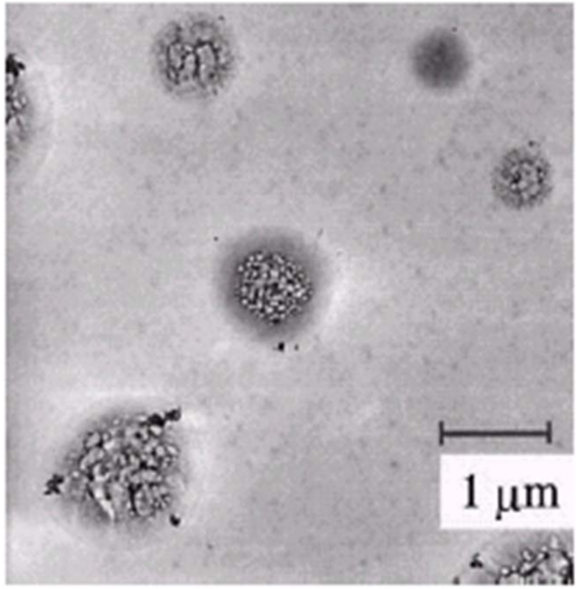

Fig. 2. Particles from LPI stage 4. Most of the particles are probably droplets that evaporated in the electron microscope. Note the soot-like aggregates that have been incorporated into the droplet. The sample was taken on May 20, 1997, on the fifth floor bridge way between Boelter Hall and Engineering IV at UCLA.

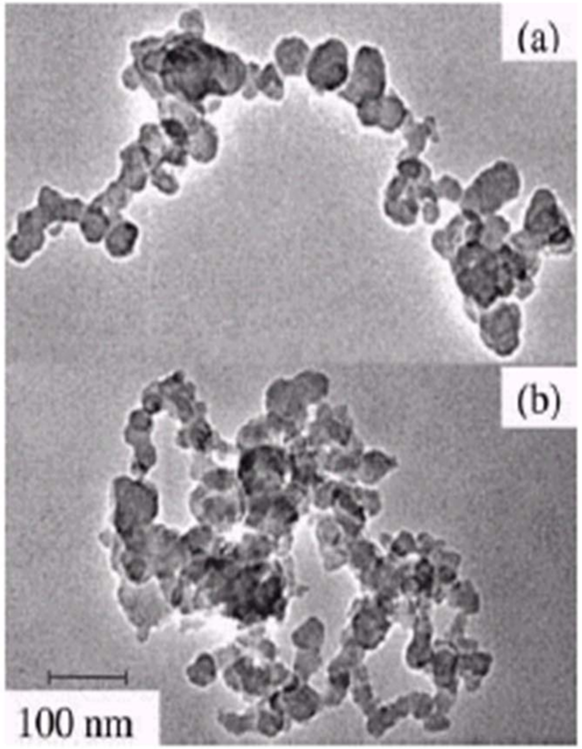

Fig. 3. Chain aggregates of ultrafine particles from stage 7. a shows a short chain with low D_f and b a longer chain with high D_f. The sample was taken on Feb 20, 2001, at the San Jacinto Air Quality Management District (AQMD) site.

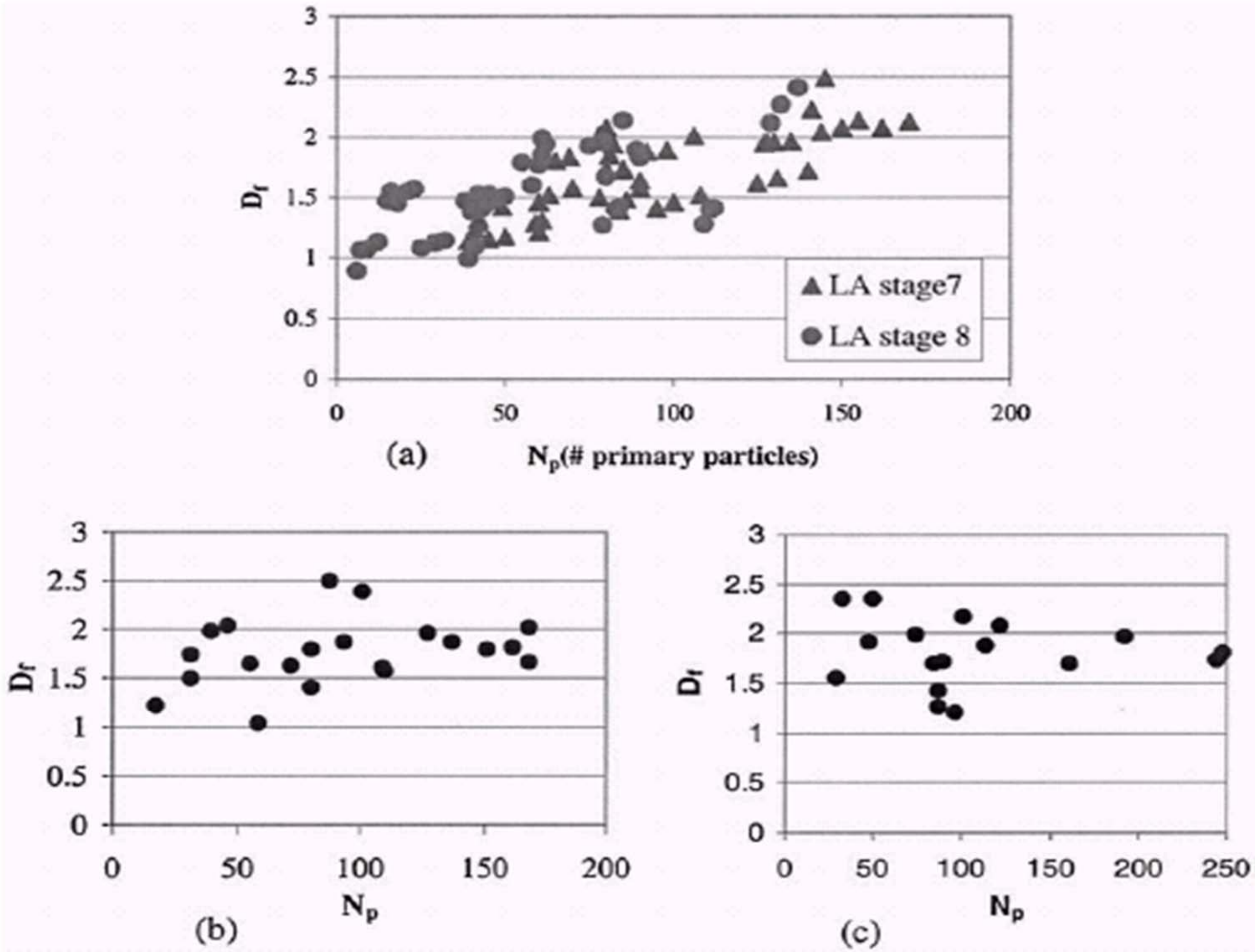

Fig. 4. (a) D_f tends to increase with N_p for particles collected over a 4-year period, 1997–2001, at five different sites: Boelter Hall–Engineering IV bridge at UCLA, Santa Monica Pier, and the Veterans Administration Hospital San Jacinto Air Quality Management District site. The average D_f is 1.63. Particles were also collected at rural areas in (b) Research Triangle Park, NC, and (c) San Jacinto, CA. The aggregates collected in North Carolina and San Jacinto had averages D_f of 1.73 and 1.80, respectively. Data for these sites did not show the same trend as the urban sites.

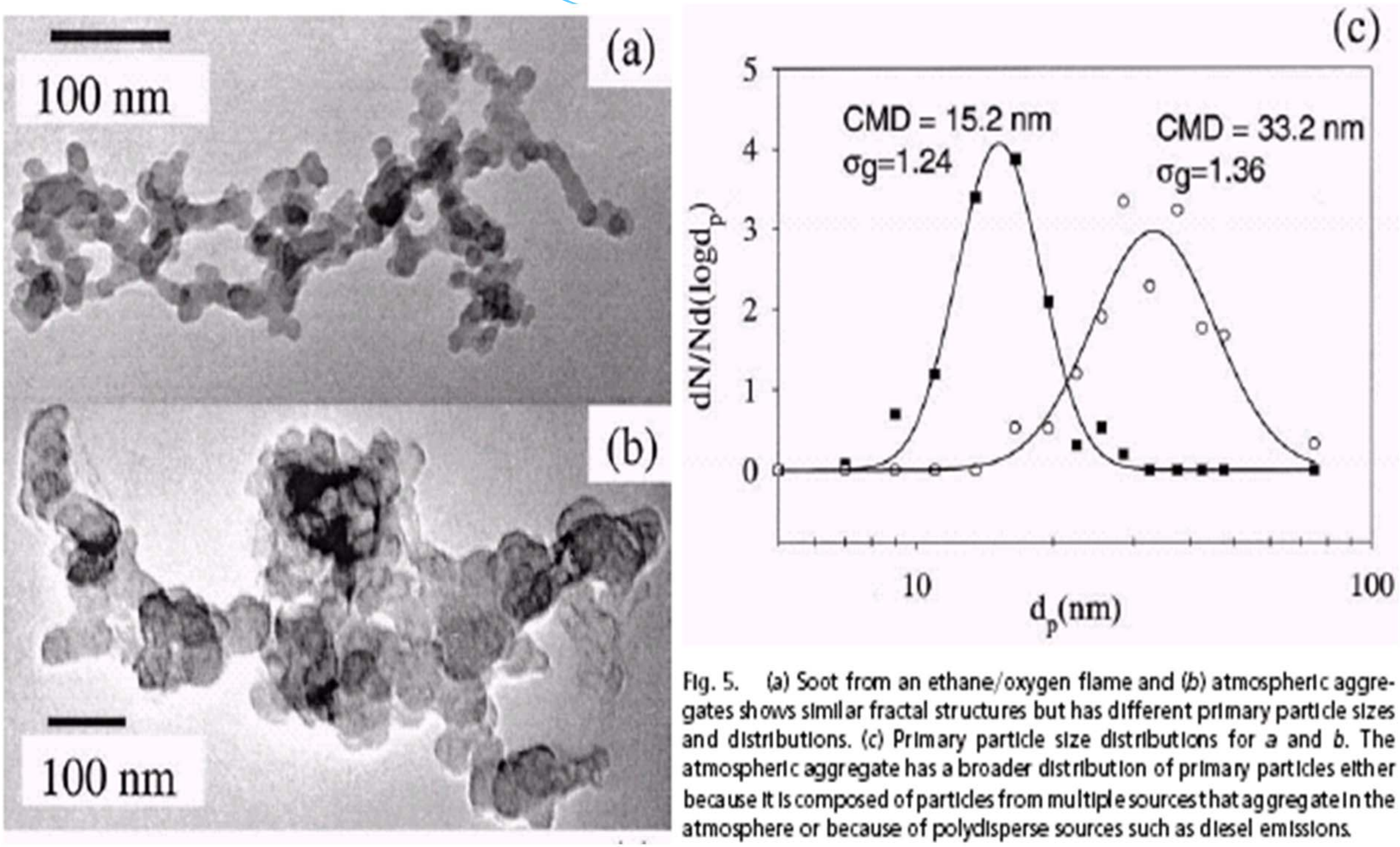

Fig. 5. (a) Soot from an ethane/oxygen flame and (b) atmospheric aggregates shows similar fractal structures but has different primary particle sizes and distributions. (c) Primary particle size distributions for a and b. The atmospheric aggregate has a broader distribution of primary particles either because it is composed of particles from multiple sources that aggregate in the atmosphere or because of polydisperse sources such as diesel emissions.

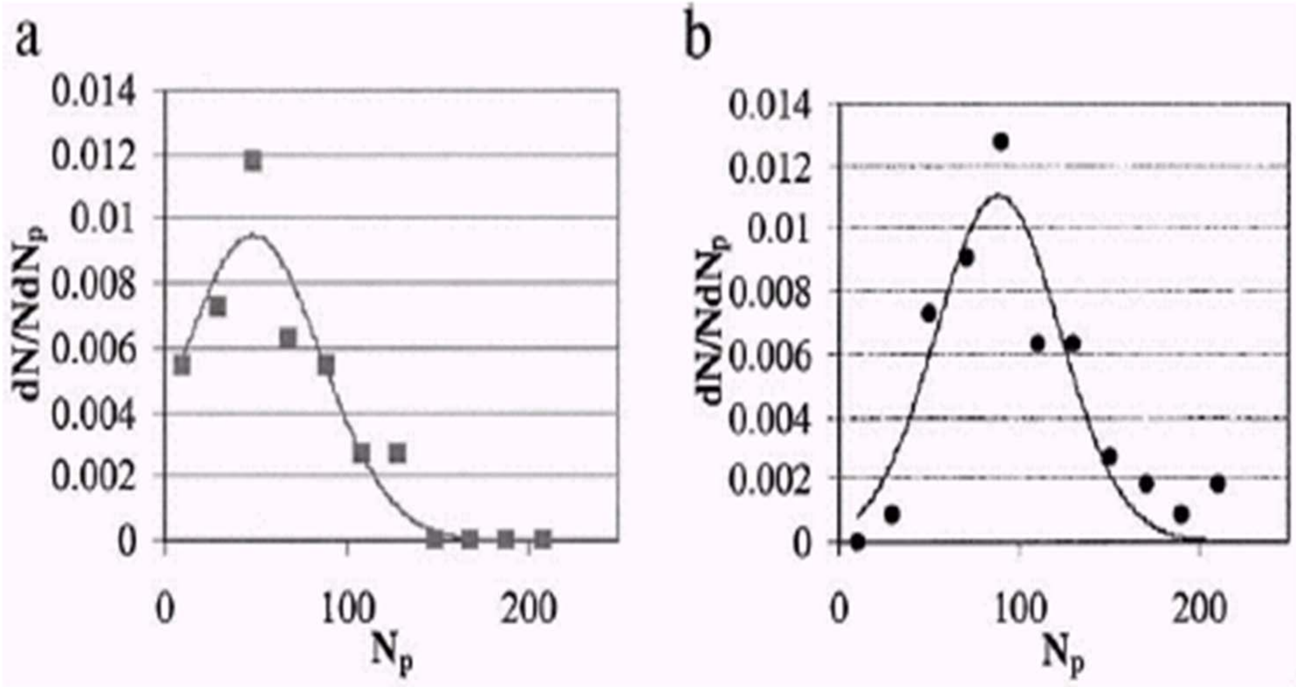

Fig. 6. Size distribution with respect to N_p of 46 and 55 aggregates for stages 7 and 8, respectively. (a) Stage 8 has a count mean N_p of 60, whereas (b) stage 7 has 120.

Conclusions

- As the size range decreases to smaller aerodynamic diameters (from 75-120 nm to 50-75 nm) the number percentage of aggregates increases from 34% to 60%.
- Atmospheric residence times, rates of transport and aggregation in atmosphere can be estimated from their data (A, D_f, and N_p).
- Polydisperse nature of atmospheric aggregates verified.
- Need more geographic disperse data for reliable estimates of large-scale effects.