Aerosol Science and Technology: History and Reviews

Edited by David S. Ensor

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About the Cover

The cover depicts an important episode in aerosol history—the Pasadena experiment and ACHEX. It includes a photograph of three of the key organizers and an illustration of a major concept of atmospheric aerosol particle size distribution. The photograph is from Chapter 8, Figure 1. The front row shows Kenneth Whitby, George Hidy, Sheldon Friedlander, and Peter Mueller; the back row shows Dale Lundgren and Josef Pich. The background figure is from Chapter 9, Figure 13, illustrating the trimodal atmospheric aerosol volume size distribution. This concept has been the basis of atmospheric aerosol research and regulation since the late 1970s.

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CHAPTER 2

Sheldon K. Friedlander

Aerosols' Man for All Seasons (1927–2007)

George M. Hidy

Most people working in the field of aerosols recognize the contributions of Sheldon Friedlander, not only from the literature but as an educator and advocate of its science establishment. This chapter traces a half-century history of Friedlander's leadership as a researcher, author, mentor, teacher, and crusader for aerosol science and technology. His achievements began in the 1950s with aerosol deposition as part of his research while an assistant professor in chemical engineering at Columbia University and later a professor at the Johns Hopkins University and the California Institute of Technology. He was recognized internationally for his aerosol science while actively committing many of his academic sabbaticals to extended visits at European institutions. His mature years in teaching and research were spent at the University of California, Los Angeles, as the Parsons Professor of Chemical Engineering and as a cofounder of the American Association for Aerosol Research (AAAR). Until his death in 2007, Sheldon Friedlander remained active as a researcher and a leader in aerosol science. He is missed for his wisdom and friendship by all who knew him.

Sheldon was an advocate of an incisive approach combining theory and experiment, following examples of Irving Langmuir and other classical physical chemists. His notable and highly original works ranged from his early study of particle deposition and diffusion and the concept of self-preservation in size distributions, to the conceptualization of chemical mass balance modeling for atmospheric aerosols, and on to recent research on the formation and growth of particles, especially in the "nano" range. He made extensive contributions to the understanding of the importance of particle size distributions for characterizing aerosol dynamics and for environmental protection. He published works on the nature and significance of secondary atmospheric aerosols as a key to managing air quality. Through two editions

of his book, *Smoke, Dust, and Haze* (Friedlander, 2000), he communicated his deep, underlying understanding and approach to aerosol science to many workers and students of the science. Perhaps most important were his philosophical principles as expressed in his education and mentoring of the many students who have followed his lead and contributed greatly to the advancement of science and technology. Given Sheldon's long-standing record of achievements, it is most appropriate to recognize him among the great pioneers of an evolving 20th-century aerosol science.

Introduction

Aerosol science and technology are largely founded on experimental measurements and observations of natural phenomena; the discipline lies somewhere in between physics, chemistry, and various areas of modern engineering. Professor Sheldon Friedlander billed aerosol science as an "enabling technology" for a wide variety of practical applications, ranging from a description of atmospheric phenomena to the prolific new world of nanotechnology. As mentor, educator, researcher, and conceptual leader, he contributed to this wide range of aerosol-relevant topics.

Sheldon's career began in the post–World War II years following the maturing of the experimental work of Victor LaMer and David Sinclair at Columbia University. Later, as a graduate student, he worked closely with H. F. Johnstone in chemical engineering at the University of Illinois. His early training in continuum fluid dynamics and chemical transport phenomena was ideally suited to advancing the knowledge about aerosol dynamics, through design of experiments and interpretation of many observations and measurements in the literature. His work began in a precomputer era, so his theoretical models relied heavily on analytical approximations and fluid boundary layer theory applied to aerosol particles rather than the blunt edge of computer modeling for problem solutions. In addition, his theoretical work was essentially always derived from or tied to experimental observations for verification of his interpretations.

As a student and colleague of Sheldon, I experienced many of the elements that made him one of the greatest aerosol scientists of our day. As a mentor, he guided many students and associates through the frontiers of aerosol science with great wisdom. His mentoring was tempered with the spice of international cuisine, especially French, and a sprinkling of fine art, including

works from the Classic and Renaissance periods, found in the J. Paul Getty museums in the Los Angeles area. As an educator, he spent a lifetime in teaching at major universities across the United States and abroad. As a researcher, he contributed extensively to the knowledge about contemporary aerosol dynamics, atmospheric aerosols, and the interface between aerosols and public health or welfare, as well as the development of new concepts in particle characterization in industrial applications. His ability to synthesize, communicate, and interpret aerosol science for students is well represented in the two editions of his book *Smoke, Dust, and Haze* (Friedlander, 2000). He also is well known for his contributions to shaping, in the 1970s and later, key principles of the US air quality management practice, the founding of the American Association for Aerosol Research (AAAR), and many cooperative projects of national and international scope, including the California Aerosol Characterization Experiment (ACHEX) (Hidy, 2011). Figure 1 depicts Sheldon in his laboratory. Other photographs of Sheldon with some of his many colleagues are included elsewhere in this book.

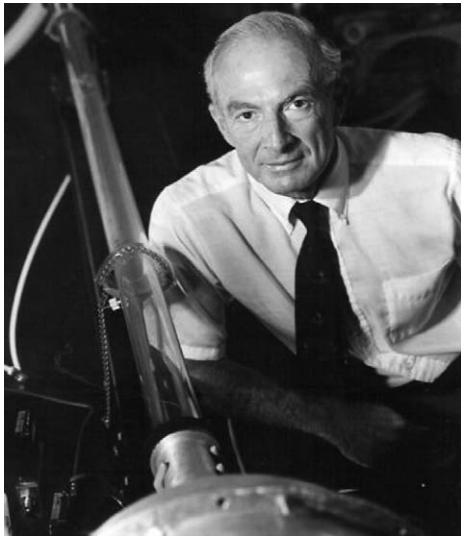

Figure 1. Sheldon Friedlander in his laboratory at the University of California, Los Angeles.
Photo: Courtesy of the Friedlander family.

As a personality, Sheldon was known to his colleagues as a true intellectual and academician. He was generally soft spoken and individualistic, with an intuition and insight akin to one of his mentors, the physical chemist Irving Langmuir. Sheldon was raised a New Yorker but spent much of his life in Southern California, with intermittent travel abroad. With his wife, Marjorie, he experienced the pleasures of family and children. As a chemical engineer

and applied scientist, he was highly intuitive in his work, with a mix of individual projects in "little" science combined with participation in larger "big" science projects. His influence in aspects of chemical engineering and aerosol science through commitments and leadership in the public domain and professional activities continue to be significant in shaping strategies for environmental protection.

Sheldon's greatest achievements will be recognized in part from his contributions to aerosol research in the many areas in which he worked with students and colleagues. I offer examples of his achievements here, mainly focusing on his work in aerosol dynamics. This tribute reflects my biases in knowledge and interpretation of his work. I hope my choice of examples will go well beyond such biases as the history of aerosol science progresses in successive generations.

Aerosol Dynamics

Sheldon's basic interest in continuum aerosol dynamics began during his initial work with Johnstone, when he investigated the deposition of aerosol particles on surfaces from a turbulent gas flow (Friedlander & Johnstone, 1957). Figure 2 shows these results, represented in dimensionless form. Sheldon's work largely extended to aerosols the concept of mass transfer from turbulent media to surfaces using boundary layer theory, with an application to pipe flow. He then extended this early work to particle transport to spheres, considering convective diffusion processes (Friedlander, 1967).

Figure 2 provides an example of a comparison between theoretical results and experiments for the Brownian diffusion and inertial capture regimes, including the results of Friedlander and Johnstone. Later Friedlander contributed to the understanding of deposition in these regimes to various geometric shapes, including spherical or cylindrical collectors as an analogy to filter fibers. The theory of diffusion and inertial deposition is commonly used in collector design and collection efficiency estimates today and is referenced in textbooks across the world. Friedlander's investigations of deposition processes for particles led to his natural interest in collision and coagulation phenomena. This interest related more broadly to formation and growth processes and his logical consideration of the general dynamic equation (GDE) for particle clouds. The formulation of the GDE basically involved a mass balance equation for particles, in the form shown in Figure 3.

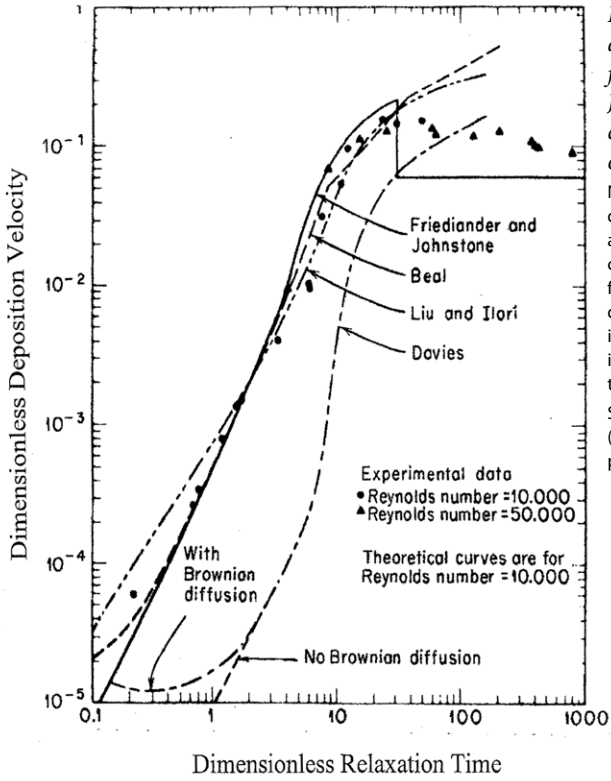

Figure 2. Turbulent deposition model from Friedlander and Johnstone (1957) in comparison with findings of other investigators.

Note: The dimensionless deposition velocity is defined as the ratio of the mass transfer coefficient k and the fluid friction velocity u^*, and the dimensionless relaxation time is given by tu^{*2}/v, where v is the kinematic viscosity of the gas.

Source: Liu and Agarwal (1974). Reprinted with permission from Elsevier.

$$\frac{dm_p}{dt} = \frac{\partial \rho_p V}{\partial t} + \rho_p \mathbf{q} \cdot \nabla V = \rho_p \int_{v_d}^{\infty} I\, dv + \rho_p (Iv)_{v_d} + \nabla^2 \int_0^{\infty} \rho_p D_p v n\, dv - \nabla \cdot \int_0^{\infty} \rho_p \mathbf{q}_F v\, dv.$$

[Growth of particles larger than a minimum of site v_d] [Formation of new particles by nucleation] [Diffusion] [Migration by external force field]

Figure 3. General dynamic equation based on a mass balance for aerosol clouds.

Note: The terms of the equation are written on the right and include advection, diffusion, sources, and action of external forces. Losses by deposition and primary sources enter through the equation's boundary conditions. Here m_p is particle mass, ρ_p is particle density, V is particle volume, q is the aerosol velocity, I is the "current" or volume of material transferred as a result of gas-to-particle conversion, v is the volume of particles in the size range v and $v + dv$, D_p is particle diffusivity, and q_F is the velocity on the particle from external forces such as those imparted, for example, by electrical fields or by gravity.

Source: Friedlander (2000).

This equation is a complex expression involving manipulation of probability distributions—size distributions—to account for the range of size that affects the particle mass accounted for. A similar form can be written to account for total mass concentration as a function of chemical composition. Researchers have constrained their investigation of the properties of the GDE to certain limiting cases, such as growth or coagulation or loss-dominated conditions. As a major part of his work, Friedlander studied in detail the attributes of elements of this equation and used them to educate students about the formalism of particle cloud dynamics. He also made use of this framework to illustrate the foundations of the dynamics equation to its various moments (integrals), which are linked with practical measurements of cumulative distributions, including filter-based determinations of mass and chemical composition of airborne particles (Friedlander, 2000).

The classical theory of coagulation is embodied in the GDE and in one simplified form involves the application of the theory of collisions by Brownian motion. As an aside, particle diffusion by Brownian motion (and by random motion in turbulence) intrigued Friedlander, especially in deriving Einstein's diffusivity from the Langevin equation. This model provided a natural extension to Smoluchowski's theory for coagulation (Chandrasekhar, 1954) as a limiting expression of the GDE.

Applying dimensional scaling to the GDE for the case of coagulation combined with sedimentation, Friedlander (1960) found that subranges of a hypothetical number-particle size distribution could be identified, which were relevant to size distributions measured in the troposphere, as reported by Junge (1963) and later determined for urban particles sampled in Baltimore (Friedlander & Pasceri, 1965). Coagulation theory subsequently led to Friedlander's application of scaling of the size distribution in terms of total number concentration and cumulative volume such that after a period of time the size distributions become uniformly the same in dimensionless form, known as *self-similar or self-preserving.* Friedlander found that the evolution of the particle size distribution to develop this form for certain processes, such as coagulation of particles in Brownian motion, could describe certain cases, such as cigarette smoke. This concept is illustrated in Figure 4, where the size distribution of the coagulating particulate cloud is shown in dimensioned form but achieves self-similarity a short period after release of the smoke.

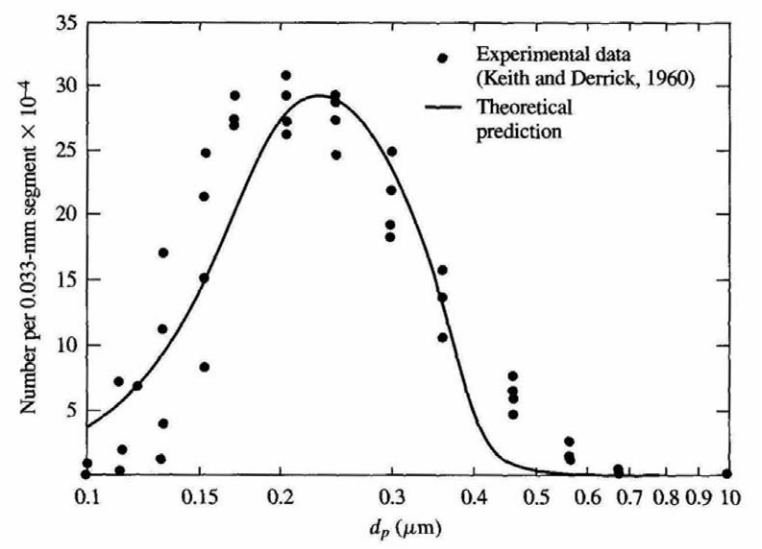

Figure 4. Comparison of experimental size distribution data for tobacco smoke with estimates based on self-preserving size spectrum theory.

Note: The volume density is 1.11 x 10^{-7}, and the total number concentration is 1.59 x 10^{7} cm^{-3}. The peak in the number distribution is estimated to be diameter (d_p) ≈ 0.2 μm.

Source: Friedlander and Hidy (1969).

Investigators have obtained other forms of self-preserving distributions by combining coagulation with growth processes. For example, Pich and colleagues (1970) found that combined coagulation by Brownian motion and particle growth from vapor condensation according to Maxwell's diffusion equation for diffusion to a sphere yielded such a distribution. I followed Friedlander and Swartz's suggestions and explored a steady state form of the GDE, including coagulation, growth, and diffusion terms with application to atmospheric aerosols (Hidy, 1972). Certain constraints exist for such applications and estimated the aging time for the aerosol size distribution to become self-preserving and in steady state. Earlier, Friedlander (1960) investigated the use of scaling as applied to atmospheric aerosols. Scaling by the total number concentration and the volume density yielded a dimensionless number-size distribution of interest. Figure 5 shows an example of this distribution for several sets of data taken in various urban and rural locations and aloft. The steady state form for self-preserving distributions conceptually is consistent with that found empirically in Figure 5. The power law regime of diameter^{-4} is often termed the *Junge subrange* (Junge, 1963),

but the extreme region representing large particles about 10 μm diameter or greater theoretically falls off as –19/4 from the action of sedimentation. Another important component of the GDE concerns particle formation and growth coagulation. The power law can be shown to decline by –5/2 (Friedlander, 1960); this fits roughly a range of 0.01–1 μm diameter.

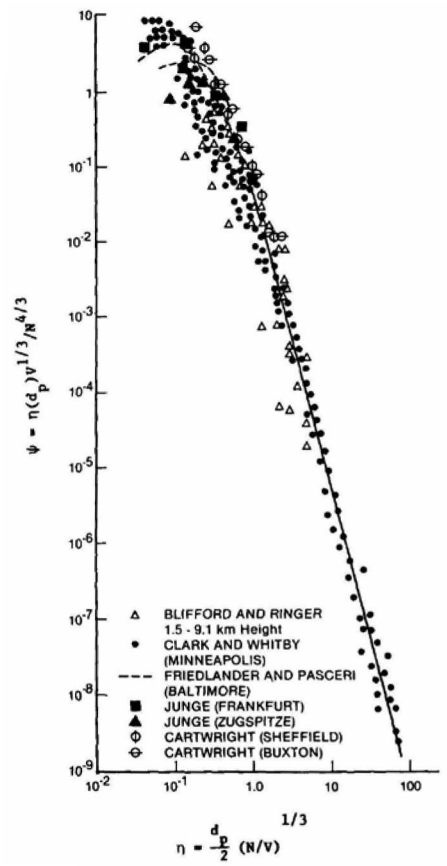

Figure 5. Dimensionless, asymptotic number-size distribution ψ for tropospheric aerosols in rural and urban conditions after Friedlander's hypothesis.

Note: Scaling is in terms of total number of particles, *N*, and cumulative volume density, *V*. Dynamic subranges are suggested in power law form. The slope of the region above η of ~2 is –4, sometimes called the Junge subrange, and the region from about 0.2 to 2 is –5/2 the coagulation subrange of Friedlander. Particle size distributions used from different sources are noted in Hidy (1986).

Source: Hidy (1986).

Friedlander's exploration of theories for particle growth by vapor condensation or chemical reactions included a review of classical theory of new particle formation by nucleation as a limiting description of molecular and particle collisions. In a more practical way, he explored the application of a vapor diffusion-condensation model to aerosol growth, noting the consistency between the continuum model and the rate of volume accumulation of particles, as illustrated in Figure 6. Here the theoretical relation between

change particle volume and mean diameter fits well the experimental data, where the theoretical line drawn through the data applies to diffusional transfer-vapor condensation, corrected for the transition from a continuum model to the free molecule regime, and including the Kelvin effect for vapor-liquid equilibrium for small droplets.

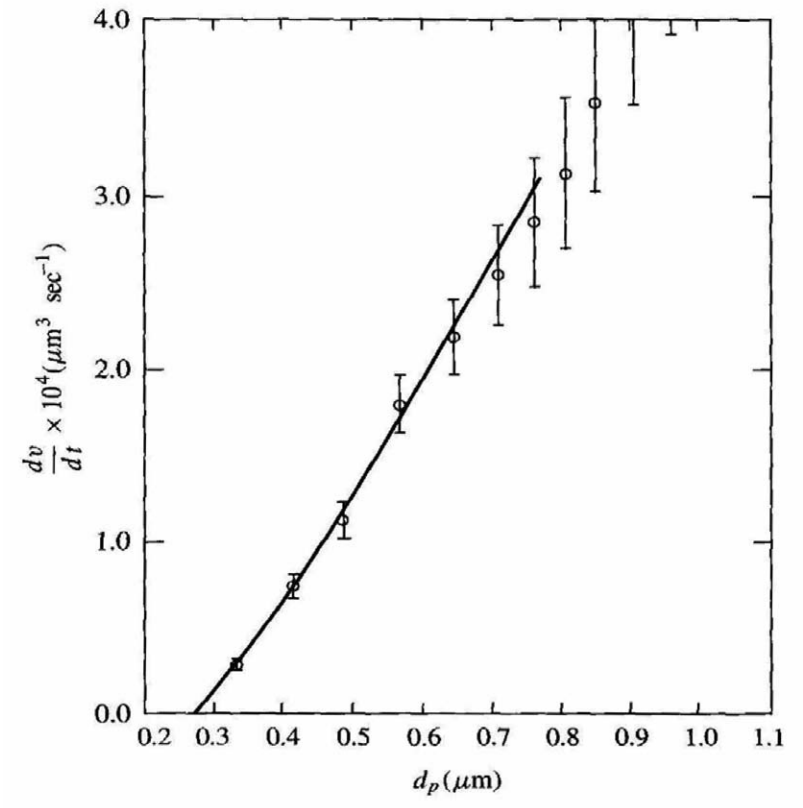

Figure 6. Growth of particles measured in a sunlight-irradiated smog chamber containing 2 ppm cyclohexene, 0.34 ppm NO, and 0.17 ppm NO_2 showing volume growth proportionality with diameter as expected from the theory of vapor diffusion and condensation on particles.

Note: The vapor diffusion model particle growth includes the Kelvin effect for droplets. The intercept of 0.27 μm is the critical droplet size.

Source: Heisler and Friedlander (1977). Reprinted with permission from Elsevier.

Atmospheric Aerosols

As noted earlier, Friedlander demonstrated a strong interest in atmospheric processes relevant to aerosols. Beyond his work on the dynamics of the particle size distribution, he contributed to knowledge of the origins and characterization of atmospheric aerosols in several ways. His interest in the formation of particle from atmospheric reactions produced observational insight into differences in mechanisms associated with sulfate production from sulfur (e.g., sulfur dioxide—SO_2) gas oxidation. Two regimes of oxidation based on collection by particle size are well illustrated in sampling in Pasadena, California, as shown in Figure 7. The first type of regime involved high relative humidity and high concentrations of sulfate and ozone.[1] Ozone represents an indicator of strong photochemical activity and the presence of high oxidant concentrations in photochemical smog, including hydrogen peroxide. High relative humidity with hygroscopic sulfate particles produces elevated liquid water content, providing a "fertile" medium for in-particle oxidation of absorbed SO_2, mainly with H_2O_2. In the second regime, of lowered relative humidity, low sulfate and oxidant concentrations evidently yields the potential for an alternate pathway of OH radical oxidation in the gas phase, and subsequent collection on particles in a broader particle size range, with maximum collection in smaller particles than the Type I case. Both mechanisms had been hypothesized, but the differentiation between the two in smog conditions was not found before this work.

The GDE also can be written in terms of mass conservation by chemical species. A simplified mass balance by chemical composition is commonly used in air pollution source-receptor modeling. Friedlander may be best known as the leading discoverer of this chemical mass balance (CMB) method for apportioning sources of aerosol particles (McMurry et al., 2004). His ideas probably originated in the early publication of Hidy and Friedlander (1970) for the case of Los Angeles. Later, Friedlander's work culminated in a more definitive paper (Friedlander, 1973).

[1] Researchers found that sulfate was a major fraction of particle samples in the Los Angeles area prior to the early 1980s and attributed it to the photochemical oxidation of SO_2 in smog before SO_2 emissions were reduced dramatically in this urban area.

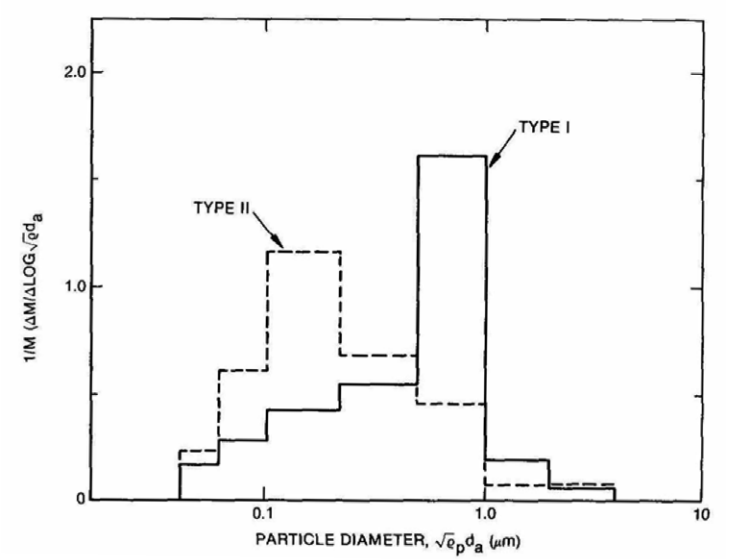

*Figure 7. Identification of two urban particulate sulfur mass (*M*) distributions with size from composite samples of several low-pressure impactor measurements in Pasadena, California.*

Note: The distributions are given in terms of aerodynamic diameter (d_a) corrected for atmospheric pressure, $\rho_P^{1/2}\, d_a$. Type I conditions include high relative humidity and high sulfate and ozone concentrations. Type II conditions involve lower relative humidity and low sulfate and ozone concentrations.

Source: Hering and Friedlander (1982). Reprinted with permission from Elsevier.

Sometime later, Gartrell and others (1980) used the ACHEX data, combined with emissions data, to develop not only a mass balance for several sites in Southern California but also a carbon mass balance for Los Angeles (Hidy et al., 1980). Table 1 presents the results of the carbon analysis. The carbon source distribution includes a large fraction from the motor vehicle sector, accounts for industrial and agricultural emissions, and provides a first comparative, empirical estimate of the primary carbon from urban sources and the secondary carbon from volatile organic compound (VOC) oxidation in the air. Analyses from the ACHEX and other investigations in California called attention to the importance of secondary organic carbon, notably by the late 1970s. It is interesting that the carbon content of the urban aerosol was largely disregarded by investigators, except in Los Angeles smog, until

Table 1. Carbon balance for smog aerosol particles at different sites in the California South Coast Air Basin (data in μg/m³)

Source	Dominguez Hills (10/5/73)	West Covina (7/24/73)	Pomona (9/6/73)	Rubidoux (9/6/73)
24-hour averages				
Motor vehicle exhaust and tire dust	1.63±0.06	5.62±0.23	3.02±0.12	2.17±0.31
Aircraft[a]	0.703±0.28	2.42±0.10	1.29±0.06	7.23±0.31
Industrial[b]	0.185±0.008	1.02±0.04	0.434±0.03	0.947±0.042
Agricultural	0	0	0	3.62±0.15
Sum-primary carbon	2.52±0.10	9.06±0.37	4.74±0.21	14.0±0.6
Total carbon	10.6±0.5[c]	24.1±1.2[c]	16.4±8[c]	15.1+0.8[c]
Secondary carbon[f]	8.08±0.51	15.0±3.0	11.7±0.8	1.10±1.00
Secondary organics[g]	12.1±0.8	22.5±2.0	17.7±2	1.65±0.8
Midday (1200–1400 hours)				
Motor vehicle exhaust and tire dust	1.92±0.087	5.83±0.24	4.54±0.17	2.24±0.89
Aircraft[a]	0.82±0.03	2.51±0.10	1.95±0.07	7.45±10.28
Industrial[b]	0.44±0.02	1.64±-0.07	1.15±0.06	1.34±0.12
Agricultural[a]	0	0	0	5.37±0.22
Sum-primary carbon	3.18±0.13	9.99±0.41	7.64±0.30	16.4±1.4
Total carbon	19.6±1.4[e]	47.6±2.4[d]	39.8±2.0[d]	20.7±1.5[e]
Secondary carbon[f]	16.4±1.4	37.6±2.4	32.2±2.0	4.26±1.7
Secondary organics[g]	24.0±2.1	56.4±3.6	48.3±3.0	6.39±2.55

[a] Assumed to be 100% carbon.

[b] Assumed to be 19% carbon.

[c] Determined for the particle less than 3–5 μm diameter; determined by conversion of C to CO_2.

[d] Total carbon conversion to CO_2.

[e] Scaled from flame ionization detector response.

[f] By difference between total measured carbon and estimated primary carbon.

[g] Assumes a relation between organic carbon and organic material of 1.5.

Note: Based on the California Aerosol Characterization Experiment (ACHEX) study (e.g., Hidy, 2006). For comparison, estimates include 24-hour averages and midday observations. The sites proceed from nominally upwind west to east from Dominguez Hills to (downwind) Rubidoux (adjacent to Riverside).

Source: Adapted from Gartrell et al. (1980).

more than a decade later, when this fraction became widely important for fine particles concentration across the United States (McMurry et al., 2004). Gartrell and colleagues (1980) also reflected Friedlander's continuing interest in growth processes by using the ACHEX data to estimate the change in the particle volume-size distribution associated with growth of particles from chemical reactions or vapor condensation. From the continuous size distributions measured in the Los Angeles area, they demonstrated the strong growth of particles in photochemical smog from chemical conversion, inferred from the CMB calculations and the size distribution data in Figure 8. These results suggested the dominance of secondary particle formation processes in urban aerosol dynamics, under the aerometric conditions of the 1970s.

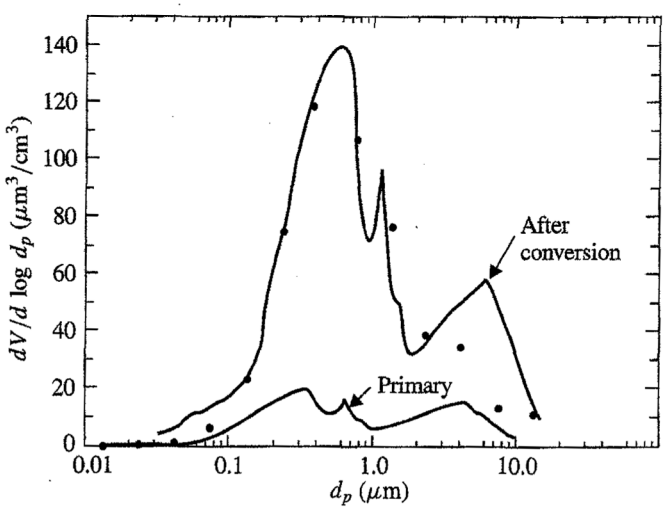

Figure 8. Volume (V)-size (diameter) distribution of primary particles estimated from material balance of Los Angeles aerosols, compared with secondary particles produced in photochemical smog during midday, October 1973.

Note: The secondary component was estimated from growth laws applied to secondary particle components (sulfate, nitrate, and carbon).

Source: Gartrell et al. (1980).

Later results reported by Turpin and Huntzicker (1995), for example, based on ambient data and emissions inventory data, showed substantially less secondary carbon formation in smog in the 1980s. For ACHEX, generally >70% secondary carbon is indicated in Table 1, whereas Turpin and Huntzicker and others reported <50% and as low as 10% secondary carbon

in Los Angeles conditions. The VOC emissions, presumed responsible for secondary particle production, may have changed composition during these two study periods, which may be a factor in the difference. The method used by Gartrell and colleagues (1980) also relied heavily on the primary particle emission inventory in the 1970s, which was probably in error, as were the data for VOC emissions. In any case, the Gartrell results strongly directed interest in the organic particle formation in smog aerosol as an important process in air pollution. This process has been studied extensively since the 1980s. Now the presence of primary and secondary organic carbon is widely recognized both in naturally occurring aerosols and in polluted air (McMurry et al., 2004).

Friedlander's interest in particulate organics took him into the area of carcinogens, represented by polycyclic aromatic hydrocarbons (PAHs) in urban air. His work with Antonio Miguel in exploring the (semivolatile) PAHs in Los Angeles led him to examine the distribution of the compounds with particle size (Venkataraman & Friedlander, 1994). These data are important for understanding the dose of PAHs that may reach the lower respiratory system in humans.

Public Health and Welfare

Although I have concentrated on the application of Friedlander's fundamental ideas to atmospheric aerosols and air pollution, his work has long been motivated by concerns for protection of human health and welfare. In addition to his work on air pollution, he investigated aspects of the deposition of particles in the human respiratory system and worked with other investigators in characterizing visibility impairment, both in the urban setting and in remote areas of the western United States. Advanced understanding of the mechanical aspects of particle deposition in the lungs as constructed from investigations of radioactive material entering the lungs provided an important link between the different modes of the ambient particle distributions and the penetration of particles sequentially into the upper and lower respiratory system. These results led to the development of a particle size–based mass concentration standard in 1987, revised in 1997 as the US National Ambient Air Quality Standard (US Environmental Protection Agency [EPA], 1996).

One of Friedlander's interesting health-related hypotheses concerned the influence of particles bearing peroxides on the human respiratory response to pollution (Friedlander & Yeh, 1998). Although this hypothesis is one of many currently of interest, it stands as a unique contribution from engineering science to the environmental medicine field.

The Ultrafine to Nanoparticle Regime

With the increasing interest in the ultrafine or the nanoparticle size range (<100 nm [0.10 μm] diameter) in the 1990s, Friedlander and his students began to investigate the properties of this extremely small material. These particle ranges have become of interest both in terms of rapidly expanding industrial applications, including the electronics industry, and in relevance to public health. In the latter case, recent studies have suggested the importance of the ambient nanoparticles for stimulating a respiratory response in humans. This hypothesis is of particular interest for combustion particles, especially those from internal combustion engines.

In any case, Friedlander began to investigate the properties of laboratory-generated nanoparticles, including refractory and crystalline materials. In addition, he explored the properties of finely divided combustion particles, identified in part as black or elemental carbon. Although these particles may be formed in the nanoparticle range during combustion or immediate entry from exhaust to the air, they can rapidly agglomerate to form chains or lumpy collections of the nanoparticles. Figure 9 shows an example of these agglomerates in comparison with apparent coalesced material. Friedlander and colleagues (1998) noted the distinction between coalesced, lumpy material composed of liquid and solid material (Figure 9a) and aggregates that form the more chain-like, loosely aggregated material (Figure 9b). The processes of agglomeration and coalescence depend on the properties of the mixed particle collections or on the surface properties of pure particles during their production. Molecular and cohesive or adhesive forces come into play, including Van der Waals forces, creating complex shapes of larger particles as they undergo change from initial formation by chemical reactions into larger particles by collision processes. This kind of research brought Friedlander from macroscopic theories of particle growth and coagulation into the world of the near-molecular processes and the application of principles from solid and liquid state physics, where a variety of potentially innovative "aerosol" engineering can be accomplished.

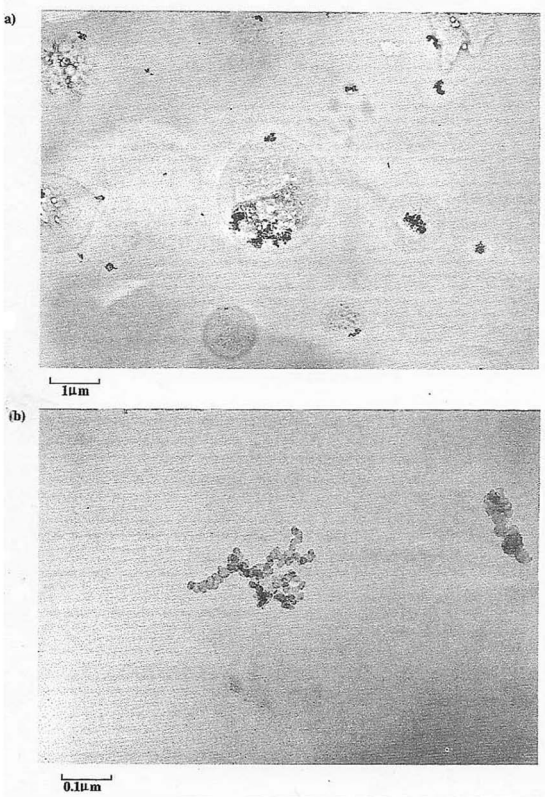

Figure 9. Electron micrographs of fine particles in the 0.1–1.0 µm diameter range, with combined solid and liquid features (a) compared with agglomerates of fine particles composed of chains of nanoparticles of ~10 nm (b).

Source: Friedlander (2000). Reprinted with permission from Oxford University Press.

Leadership, Mentoring, Interactions, and Teaching

Friedlander's role in mentoring and teaching represents an important part of his life in aerosol science and chemical engineering. Aside from his faculty contributions to intellectual life at Columbia's School of Engineering, the Johns Hopkins University, the California Institute of Technology, and, last but not least, the University of California, Los Angeles, he has guided or exchanged thoughts and ideas with researchers and students throughout the world of aerosols since the 1950s. Among the students who have benefited from Sheldon's wisdom are E. Avol, B. Dahneke, S. Ehrman, G. Gartrell, S. Heisler, S. Hering, G. Hidy, R. Husar, H. Jang, A. Kao, K. Keller, F. Lai, E. Macias, P. McMurry, A. Miguel, M. Miller, R. Pasceri, S. Pratsinis, P. Roberts, A. Stelson, D. Swift, C. Venkataraman, C. Wang, A. Weber, W. White, and many others. Many of these contemporaries have become well known in their own right. His connection with the health sciences is represented by the work of former students K. Keller, R. Pasceri, and D. Swift.

Among the colleagues that worked or interacted closely with Sheldon and mutually enriched their world of aerosols are D. Allen, P. Altshuller, J. Bricard, G. Cass, R. Charlson, J. Chow, W. Clark, F. Fissan, R. Flagan, N. Fuchs, A. Goetz, D. Grosjean, A. Haagen-Smit, J. Hales, G. Israel, C. Junge, M. Kerker, M. Lippmann, B. Y. H. Liu, J. Lodge, M. Madelaine, R. McClellan, P. Mueller, N. Nelson, J. Pich, W. Pierson, J. Pitts, O. Preining, D. Pui, J. Seinfeld, G. Sem, A. Serafin, K. Spurny, W. Stober, G. Sverdrup, J. Watson, K. Whitby, and W. Wilson. Just this partial list indicates well Sheldon's broad interest in challenging his colleagues and associates to address the many complexities of aerosol science and applications. These challenges have inspired the leaders and their students in aerosol science, in combination with new instruments and theories, to advance the state of knowledge farther in the last 50 years than centuries previously.

Friedlander's recognition and achievements are broadly known among his peers, including his election to the National Academy of Engineering and his many awards for scholarly excellence from the American Institute of Chemical Engineers, AAAR, and AAAR's international counterparts. Possibly best known is his public service as chairman of the Clean Air Science Advisory Committee in the 1980s and as an advisor to the National Institute of Environmental Health Sciences. Equally important was his key role in founding the AAAR and his subsequent organization of analogous international associations.

His commitment to cooperative science in aerosols was well exemplified early on with the Pasadena experiment in 1969, which provided pioneering data characterizing photochemical smog in Southern California (Hidy, 2006). This ad hoc summer campaign led to the ACHEX, in which he was a co-principal investigator. The ACHEX, sponsored by the California Air Resources Board, was one of the first, if not the first, major cooperative experiment addressing in the 1970s the characterization of natural baseline and air pollution aerosols, with focus on photochemical particle formation (Hidy et al., 1980; Hidy, 2011).

His sponsored research began in the 1950s with grants from the US Atomic Energy Commission and the National Science Foundation. Later, in the 1980s, Friedlander formed a center for environmental research at the University of California, Los Angeles, to create for EPA and other sponsors a vehicle for cooperative, multidisciplinary programs to address contemporary environmental issues in air quality, waste management, and environmental

risk assessment. His advocacy for innovation in aerosol science and technology continues today with international lectureships describing the field as the "enabling science and technology" for future applications of fine particles into the nano size range.

Conclusion

Sheldon Friedlander's contributions to the knowledge about aerosol properties and dynamics, especially in relation to the fundamentals of particle deposition, formation, and growth processes, led more than 50 years of rapid progress in this field. The application of his work to environmental protection has led to pioneering insight into fine particles, with the goal of improving air quality. Friedlander's continuing productivity as a researcher in engineering science showed a creativity and insight rare among his peers. In addition to his research contributions, he was an exceptional leader in his achievements as a mentor, educator, and advocate for aerosol science and technology. Until his death in 2007, his contributions and worldwide recognition among his colleagues ranked him as one of the greatest living aerosol scientists of our time.

References

Chandrasekhar, S. (1954). Stochastic problems in physics and astronomy. In N. Wax (Ed.), *Selected papers on noise and stochastic processes* (pp. 3–91). New York, NY: Dover.

Friedlander, S. K. (1960). Similarity considerations for the particle-size spectrum of coagulating-sedimenting aerosol. *Journal of Meteorology, 17,* 479–483.

Friedlander, S. K. (1967). Particle diffusion in low speed flows. *Journal of Colloid and Interface Science, 23,* 157–164.

Friedlander, S. K. (1973). Chemical element balances and identification of air pollution sources. *Environmental Science and Technology, 7,* 235–240.

Friedlander, S. K. (2000). *Smoke, dust, and haze: Fundamentals of aerosol dynamics* (2nd ed.). New York, NY: Oxford University Press.

Friedlander, S. K., & Hidy, G. M. (1969). New concepts in aerosol size spectrum theory. In *Proceedings of the Seventh International Conference on Condensation and Ice Nuclei.* Prague: Czechoslovak Academy of Sciences.

Friedlander, S. K., Jang, H. D., & Ryu, D. H. (1998). Elastic behavior of nanoparticle chain aggregates. *Applied Physics Letters, 72,* 173–176.

Friedlander, S. K., & Johnstone, H. F. (1957). Deposition of suspended particles from turbulent gas streams. *Industrial and Engineering Chemistry Research, 49,* 1151–1156.

Friedlander, S. K., & Pasceri, R. (1965). Measurement of particle size distributions of atmospheric aerosols. II. Experimental results and discussion. *Journal of the Atmospheric Sciences, 22,* 577–584.

Friedlander, S. K., & Yeh, E. K. (1998). The submicron atmospheric aerosol as a carrier of reactive chemical species: Case of peroxides. *Applied Occupational and Environmental Hygiene, 13,* 416–420.

Gartrell, G., Jr., Heisler, S. L., & Friedlander, S. K. (1980). Relating particulate properties to sources: The results of the California Aerosol Characterization Experiment. In G. M. Hidy (Ed.), *The character and origins of smog aerosols* (pp. 665–713). New York, NY: Wiley Interscience.

Heisler, S. L., & Friedlander, S. K. (1977). Gas-to-particle conversion in photochemical smog: Growth laws and mechanisms for organics. *Atmospheric Environment, 11,* 158–168.

Hering, S., & Friedlander, S. K. (1982). Origins of aerosol sulfur size distributions in the Los Angeles basin. *Atmospheric Environment, 16,* 2647–2656.

Hidy, G. M. (1972). The dynamics of aerosols in the lower troposphere. In T. Mercer, P. Morrow, & W. Stober (Eds.), *Assessment of airborne particles: Fundamentals, applications, and implications to inhalation toxicology* (pp. 81–115). Springfield, IL: Charles C. Thomas.

Hidy, G. M. (1986). Definition and characterization of suspended particles in ambient air. In S. T. Lee, T. Schneider, L. Grant, & P. J. Verkerk (Eds.), *Aerosols: Research, risk assessment and control strategies* (pp. 19–41). Chelsea, MI: Lewis.

Hidy, G. M. (2011). Reminiscences about Pasadena, ACHEX, and beyond. In David S. Ensor (Ed.), *Aerosol Science and Technology: History and Reviews* (Chapter 8). Research Triangle Park, NC: RTI Press.

Hidy, G. M., & Friedlander, S. K. (1970). The nature of Los Angeles aerosol. In H. England (Ed.), *Proceedings of the 2nd IUAAPA Clean Air Congress.* New York, NY: Academic Press.

Hidy, G. M., Mueller, P. K., Grosjean, D., Appel, B. R., & Wesolowski, J. J. (1980). *The character and origins of smog aerosol* (pp. 1–17). New York, NY: Wiley Interscience.

Junge, C. E. (1963). *Air chemistry and radioactivity.* New York, NY: Academic Press.

Keith, C. H., & Derrick, J. E. (1960). Measurements of the particle size distribution and concentration of cigarette smoke by the "conifuge." *Journal of Colloid Science, 15,* 340–347.

Liu, B. Y. H., & Agarwal, J. K. (1974). Experimental observations of aerosol deposition in turbulent flow. *Journal of Aerosol Science, 5,* 145–148.

McMurry, P., Shepherd, M., & Vickery, J. (Eds.). (2004). *Particulate matter science for policy makers: A NARSTO assessment.* Cambridge: Cambridge University Press.

Pich, J., Friedlander, S. K., & Lai, F. S. (1970). The self preserving particle size distribution for coagulation in Brownian motion-III. Smoluchowski coagulation and simultaneous Maxwellian condensation. *Journal of Aerosol Science, 1,* 115–126.

Turpin, B. J., & Huntzicker, J. J. (1995). Identification of secondary organic aerosol episodes and quantitation of primary and secondary organic aerosol concentrations during SCAQS. *Atmospheric Environment, 29,* 3527–3544.

US Environmental Protection Agency. (1996). *Air quality criteria for particulate matter* (EPA/600/P95/001a, F, Vol. I–III). Research Triangle Park, NC: US Environmental Protection Agency, Office of Research and Development.

Venkataraman, C., & Friedlander, S. K. (1994). Size distribution of polycyclic aromatic hydrocarbons and elemental carbon. 2. Ambient measurements and effects of atmospheric processes. *Environmental Science and Technology, 28,* 369–572.