Atmospheric Aerosols: Some Highlights and Highlighters, 1950 to 2018

G. M. Hidy¹

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Abstract

The science of atmospheric aerosols began more than a century ago; it has experienced major advancements after the mid-twentieth century with motivation from diverse public interests and concerns for environmental protection. At least six generations of mentored investigators have involvement in these advancements. Since the 1950s, important knowledge has emerged in the theory of the dynamics of suspended particles and advanced measurements. Important developments in the theory of atmospheric aerosols include: (a) nucleation and growth mechanisms, (b) formalization of particle dynamics in the Knudsen regimes, (c) characterization of the mechanisms for the particle-size distribution, (d) identification of chemical processes for atmospheric particle sources, and (e) model integration of particle physical and chemical processes with meteorological processes. Important advances in measurements have included: (a) semicontinuous determination of particle-size distributions, (b) new methods for sampling and analysis of mass concentration and composition, (c) methods for continuous characterization of aerosol chemical properties, and (d) development of direct sensing techniques using optical properties. Examples of breakthroughs in these areas are given in the text. Illustrations of achievements in each of the areas are included in the paper. The survey is completed with comments on the generational nature of investigator contributions to aerosol science.

Keywords Aerosol history · Aerosol physics · Aerosol chemistry · Atmospheric aerosols

1 Introduction

Smoke and haze have resulted from human activity for centuries. Their presence was exacerbated with the industrial revolution and urbanization. Observations of the atmosphere have shown varying visual impairment from smoke plumes and respiratory stress accompanying haze (Brimblecombe 1987; Husar 2000). Scientific characterization of airborne particles began to emerge in the nineteenth century with knowledge of cloud formation from supersaturated vapors and optical physics. The application of fluid mechanical principles governing forces acting on small spheres and their interactions with each other or vapors set the stage for

developing aerosol physics. The “mysterious” origins of falling dust and pervasive gray-brown haze led investigators to study these phenomena. Once collected on a substrate, light microscopy could reveal properties of particles by size to $> 0.5 \mu\text{m}$ diameter. In the early-to-mid-twentieth, century aerosol mechanics continued to develop along with advanced measurement techniques for in situ observation and sampling (Podzimek 2000). With evolving modern measurement methods and motivation to improve air quality, knowledge of atmospheric aerosols has literally exploded into the early twenty-first century.

The nineteenth century aerosol-related science expanded in the twentieth century to include advances in theoretical models, experimental tools, and observations of atmospheric aerosols. Early summaries of aerosol measurements and properties are found in Whytlaw-Gray and Patterson (1932) and Green and Lane (1957). Junge (1963) summarized the mid-20th Century knowledge of atmospheric aerosols based on observations and application of theoretical constructs to interpret the evolution of size distributions, nucleation, and gas–particle interactions. Junge’s pioneering summarization

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✉ G. M. Hidy
ghidy113@gmail.com

¹ Envair/Aerochem, Placitas, NM, USA

largely anticipated the rapid growth in the science of atmospheric aerosols seen after the mid-twentieth century.

Atmospheric aerosol science has relied heavily on observations for answers to the questions, “What’s there and how did it get there”. However, beyond cataloging the nature of atmospheric aerosols, scientists have relied on the fundamentals of aerosol science for interpretation of the atmospheric particle characteristics. The main features of the science applicable to atmospheric particles became well established by the 1990s. The theoretical components of aerosol science of interest focused on mechanics and chemical mechanisms. The experimental or observational component involved development and use of unique sampling and physicochemical tools combined with application of advancing meteorological knowledge affecting particle behavior.

The development of continuum theory for fluids provided a basis for characterizing particle motion according to conditions investigated in Stokes’ work (e.g., Lamb 1932) and later for heat and mass transfer to suspended objects (e.g., Byrd et al. 1960). Alternatively, statistical mechanics and the Boltzmann Equation supplied a framework for aerosol dynamics, formalized for gasses (e.g., Chapman and Cowling 1960) and later for aerosols (Hidy and Brock 1970).

The theory of aerosol mechanics using continuum theory for single particles and for clouds of particles evolved to include accounting for “slip” at a particle surface and inertial effects vs. the consequences of Brownian motion of very small particles. The former emerged from Millikan’s (1913) experiments of falling droplets. The latter was discussed in Einstein’s (1905) paper on diffusion, and later was contained in Smoluchowski’s (1917) theory of coagulation. The status of aerosol mechanics was well described by Fuchs (1964), and later in texts such as Hidy (1984), Friedlander (1990), and Seinfeld and Pandis (1998).

Early observations of atmospheric aerosols derived from study of optical effects and the thermodynamics of phase change. Key optical effects associated with light scattering from suspended particles emerged in theoretical solutions of Maxwell’s equation by Rayleigh 1871 for small particles and for larger particles (Lorenz 1890; Mie 1908). Tyndall’s experiments on color from particle scattering were reported in 1869. The classical optical theories were advanced for aerosol light scattering and absorption well into the mid-twentieth century (e.g., Kerker 1969). Optical properties play a key role in radiative transfer for visibility theory and for describing the earth’s thermal radiation balance (Chandrasekhar 1960).

The role of airborne particles as nuclei for atmospheric hydrometeor formation developed beyond nineteenth century concepts of nucleation (e.g., Mason 1957). Cloud-forming processes in expanding air volumes was the basis for Wilson’s cloud chamber experiments in the 1890s—leading to an understanding of the role of supersaturation

in particle formation as homogenous nucleation compared with ion or heterogeneous nucleation from cosmic ray penetration (Mason 1957). Wilson’s (1897) experiments were anticipated by Aitken’s (1887/1889) studies of atmospheric nuclei and atmospheric cloud formation. The presence of Aitken nuclei in the atmosphere was reviewed extensively in Landsberg’s (1934) work.

The theories of chemical evolution of atmospheric particles were a subset of expanding knowledge of gas-phase photochemistry (Leighton 1961). Characterization of sources included soil dust, sea spray, oxidation of natural hydrocarbons, and anthropogenic emissions from industry, material and energy production and use, and commercial and residential activity (e.g., Hidy 1984). Junge’s (1963) descriptions reflect how workers recognized early on that particle sources in the atmosphere were distributed over a wide range of sizes depending on the formation, agglomeration or disaggregation processes involved. Chemical distinction by size emerged in the 1960s from size fractionated sampling and theoretical considerations describing particle formation.

Observations of atmospheric aerosols used a range of instrumentation applying various means of collection on microscope substrates, filter media, impactors, and diffusion batteries. Continuous observations were made using a number of techniques including nuclei counters, ion counters, electrical mobility analyzers, and optical instruments for acquisition of light extinction coefficients and optical density. After World War II, scientists extended their observations from the ground through the troposphere well into the stratosphere using aircraft and balloon sampling.

Collection of atmospheric particles for laboratory analysis began in the 1930s, but developed more rapidly after the 1960s. Sampling by filtration followed by laboratory analysis of water extracted material or total elemental determinations by atomic absorption spectroscopy or other detectors is exemplified in Lodge (1988) and Spurny’s (1999) books. Advances in micro-chemical analyzes for inorganic and organic materials resulted in their widespread application for bulk chemical characterization and for seeking relationships between sources and ambient composition (e.g., chemical mass balance (Friedlander 1973; Watson 1979; Watson et al. 1984) and factor analysis (Hopke 1985) solutions (Watson et al. 2016).

The development of physical and chemical methods for characterizing aerosols has facilitated a major and sustained effort worldwide to create knowledge of ambient aerosol concentrations and sources from the Earth’s surface well into the stratosphere and beyond. The collection and cataloging of atmospheric aerosols have created large data collections, whose recording and manipulation have depended on the emergence of computer data acquisition and storage technology as an adjunct to contemporary instrumentation applications.

As workers in this specialized field of science, it is useful to remind ourselves of the state of science and those who have led the way to this state. To this end, this is an “old engineer’s” survey influenced by my bias for the work US investigators. It begins with important progress in applying the basic principles characterizing atmospheric aerosols, and identifies along the way many of the scientists who made this progress possible. As a part a 21st Century portfolio of knowledge, a view of measurements characterizing aerosols in the atmosphere is then presented. The latter includes both physical and chemical measurements quantifying particles from earth’s surface to the stratosphere. A theme that runs through the narrative is a “commonly known” generational evolution of the science with exploration of atmospheric aerosols linked through institutional and professorial mentoring relationships. Like much of the study of natural science, atmospheric aerosol research has been supported mainly by government resources in the public interest of environmental protection.

2 Theory

2.1 Physical Models

Through the plethora of research of atmospheric aerosols since the 1950s, a number of major achievements can be identified relative to previous work. These include: (a) developing nucleation and growth theory for homogeneous and heterogeneous systems; (b) formalizing the dynamics of particles over a wide range to include continuum to free molecule behavior; (c) developing a theory for the aerosol size distributions; (d) identifying chemical processes forming inorganic and organic particles in the atmosphere; and (e) integrating sources, and physical and chemical processes to formulate models to estimate spatial and temporal patterns of particle characteristics. The following is a summary of these contributions beginning with nucleation phenomena. In each of the chosen topics, references to the work of groups of researchers are noted, thereby illustrating their generational origins.

2.2 Nucleation

Nucleation begins the formation of particle-size distributions, addressing new particle production in a transient atmosphere near molecular size range. Attention to the Knudsen regimes completes the mechanical theory of aerosols in the atmosphere. Theoretical interpretation of the particle-size distribution identifies key mechanical and chemical processes dominating parts of the observed in the distribution’s very broad range of particle size and number concentration. Identifying mechanisms for particle formation elucidates the complexities of chemistry of inorganic

and organic species in relation with atmospheric gasses. In addition, the process integration by modeling is crucial to interpretation of atmospheric aerosol origins and evolution by linking sources of particles with the microscale processes affecting aerosols with macroscale phenomena associated with meteorological phenomena.

The phenomenon of particle nucleation from a super-saturated gas emerged from kinetic theory, for example, in the Becker and Doring (1935) analysis, further interpreted in Frenkel (1955). This process requires a relatively high vapor supersaturation that could be obtained in a gas expansion chamber, in moist hot jets mixing with a cool gas, or in supersonic flow nozzles. Meteorologists estimated that atmospheric water supersaturation to form clouds was only a few percent (e.g., Pruppacher and Klett 1978). Study of these extremes conceptually accounted for spontaneous particle formation to concepts of atmospheric nucleation depending on heterogeneous processes in the presence of ions or small aerosol particles.

A series of improvements in the theory of homogeneous nucleation in gas mixtures took place after the 1950 (e.g., Reiss 1950; Mirabel and Katz 1974) with indirect applicability to the atmosphere. The universal presence of particles, including ions, has caused questioning of the relevance of homogeneous nucleation owing to difficulty in achieving high supersaturations in the atmosphere. Ions were known to investigators by the turn of the twentieth century. Israel and Schulz (1932), Nolan et al. (1925) and Nolan and Doherty (1950) discussed aspects of their presence. Junge (1952, 1963) and Pruppacher and Klett (1978) summarized the knowledge of ion nucleation and aerosol particles. Research on the theory of ion nucleation has continued into the 1990s and beyond as part of the study of $\sim 0.01 \mu\text{m}$ diameter particles. Theoretical conditions for ion-mediated nucleation are discussed by Enghoff and Svensmark (2008) and later Yu (2010), who have reviewed the current state of knowledge about ions and nucleation in the atmosphere.

In the 1980s, aerosol observations in remote areas suggested that homogenous nucleation could be relevant to production of particles $< 0.01 \mu\text{m}$ diameter (e.g., Weber et al. 1997; Kulmala et al. 2003). This led to a resurgence of theoretical studies that investigated sulfuric acid nucleation in the presence of ammonia and water, and the possibility for achieving conditions for the formation of low volatility organic species by this process (Zhang et al. 2012).

2.3 Rarefied Gas Dynamics

The nucleation process represents only one of a number of key areas extending aerosol dynamics into the ultrafine particle range less than $0.05 \mu\text{m}$ diameter. This direction also was expanded through application of concepts of rarefied gas dynamics, combining approximate solutions for the Boltzmann

equation and from continuum mechanics. Semiempirical theory for small particle dynamics was discussed by Fuchs (1964) originally published in Russian in 1955. He summarized a variety of experimental and theoretical work from the early to mid-twentieth century on ultrafine particles including molecular clusters. The aerosol dynamics relationships from free molecule flow through a transition regime to continuum flow were further codified in Hidy and Brock (1970). The principal parameter for this relationship is the Knudsen number (Kn), the ratio of the mean free path of the suspending gas-to-particle radius.

Particles $< 1 \mu\text{m}$ diameter in the atmosphere are representative of dynamics in the transition regime $10^{-2} < Kn < 1$ near the earth's surface, and move towards free molecule behavior at higher altitudes for $Kn < \sim 10$. The change in dynamical theory was not widely recognized by aerosol scientists in their conceptual models well into the 1950s. The regimes for the lower troposphere are illustrated schematically with Junge's form of the particle-size distribution in Fig. 1. Here, the free molecule regime nucleation kinetics were for particles $< \sim 0.01 \mu\text{m}$ radius in the lower troposphere. With altitude the Knudsen regimes increasingly "influence", the dynamics of larger particles as the mean free path of the suspending gas increases.

The theory for the Knudsen dynamical regimes has been refined since the 1960s, extending the theoretical models from that time to specific cases of interest. Examples indicate continued interest in refining the theories of momentum and mass transfer. Sangers et al. (2018) discuss drag in nearly free molecule flow; Carson et al. (2017) re-examine the friction factor for aggregates in the Knudsen range. Rogak et al. (1993) investigated mobility and aggregate structure. Mass transfer as evaporation is addressed by Wanguang and Davis (1996), whereas Lushnikov et al. (1996) have looked at enhanced condensation of vapors in the transition regime. Particle growth in the free molecule and transition regimes has been studied by Tsang and Hippe (1988), Koutzeugogi et al. (1996), and Thajudeen et al. (2015). While these examples from the contemporary literature are hardly exhaustive, they do indicate that researchers continue to report progress on aerosol theories in the Knudsen regimes. Especially useful are the particle cloud dynamic calculations from numerical solutions of the "general dynamic equation (GDE)." For cases of coagulation, nucleation, and condensation growth, a limitation of theoretical applications were reported in the 1960s (e.g., Hidy 1984; Friedlander 1990).

2.4 Size Distributions

The third area of contemporary theory derives from the approximations for solutions to the GDE, and concerns the evolution of atmospheric aerosol particle-size distributions. These distributions, including number size, surface area size,

Fig. 1 Conceptual diagram of Knudsen regimes for Junge's particle number-size (radius) distribution in the lower troposphere. As visualized in the 1960s the nucleation regime below $10^{-2} \mu\text{m}$ radius is shown as intermittent because of the short residence time of these small particles. (From Hidy and Brock 1970; courtesy of Elsevier)

and volume (mass) size, are fundamental to the characterization of atmospheric aerosols. Extensive measurements used different techniques prior to the 1960s including diffusion batteries, electric mobility, light scattering, and inertial impaction (e.g., Junge 1963). Measurements led to empirical knowledge of the form of aerosol particle distributions. The number-size distributions synthesized cover an enormous range of particle size and number, as suggested in Fig. 1. The theory for the evolution of particle distributions began with Junge's considerations for nucleation, coagulation, and sedimentation of particles. Remarkably, Junge's work observationally showed that the form of size distributions is similar at different locations in the troposphere and the stratosphere.

Guided by Junge's work, the particle dynamics in shaping the size distribution were analyzed in detail in Friedlander's (1960, 1990) investigations, wherein he noted the asymptotic nature of the measured number distributions with subranges including coagulation and sedimentation, inertial collision, and vapor growth and nucleation. His research with several colleagues led to fluid dynamic, similarity theory, and the concept of self-preserving distributions that evolve in certain limiting cases such as coagulation by Brownian motion

and by condensation and coagulation growth. An example of a “universal” non-dimensional form of the number-size distribution can be shown from scaling of a variety of size distributions measured at different locations (Fig. 2).

An alternative approach to characterizing particle-size distributions assumes a form of the distribution and its moments. This approach was adopted by Whitby et al. (1972) from measurements (see also next section). Measurements were fit to a log-normal distribution form, and showed “modes” beginning with a multimodal (often bimodal or trimodal) form for volume or surface size distributions from remote or rural areas. These merge into tri- or multimodal distributions for urban areas. The modal separation follows from arguments relating to dynamic regimes. This separation was applied to health effects studies (e.g., lung deposition, Lippman and Altshuler 1975) and created a basis for regulatory focus on fine particles less than 2.5 μm diameter (Wilson and Suh 1997). Jaenicke (1986) also discussed size distributions as key functions for tropospheric aerosol physics and chemistry.

Conceptually, the modes seen in the volume distribution, for example, are strongly influenced by sedimentation at the largest particles (>5 μm diameter); the accumulation mode (0.1–5 μm), where sedimentation, inertial collisions, and growth rates are relatively low, and the nucleation and growth mode (<0.1 μm), where condensation, coagulation, and diffusion are prevalent processes. Growth of particles from vapor condensation on existing particles (heterogeneous nucleation) or chemical reactions is important; semiempirical growth laws have been identified for the submicrometer regime (e.g., McMurry and Wilson 1982).

The theory for the evolution of particle-size distributions follows from approximate solutions to the GDE. A number of publications after the 1980s that give approximate solutions to the GDE for various assumptions of the form of the collision coefficient and the nature of the nucleation, diffusion or growth processes. Examples of these approximate solutions are discussed in Toon et al. (1988); Kalani and Christofides (2002), Yu and Luo (2009), Zaveri et al. (2010) and Liu et al. (2018).

Fig. 2 Normalized form of atmospheric particle number size distribution scaled according to the “similarity” model of Friedlander (1960). $\Psi = n(d)V^{1/3}/N^{4/3}$ and $\eta = d(N/V)^{1/3}$, where $n(d)$ is the number of particles per unit volume in the diameter range $d+\Delta d$, V is the volume concentration, and N is the number concentration. The slope of d^{-4}

for larger scaled particle diameters suggests a power law form for a range of diameters qualitatively consistent with a coagulation-sedimentation process. (From Hidy et al. 2013; courtesy of Taylor and Francis)

2.5 Particle Chemistry

The complexity of physical theory for particle dynamics is compounded for atmospheric aerosols considering their chemical composition by size. By the beginnings of the twentieth century, the public understood that atmospheric particles as smoke, dust, and mists could derive from combustion, industrial activities (including metals' production), suspended soil or road dust, sea salt, volcanic eruptions, and even a dash of micrometeorites from space. The quantitative details of bulk chemical composition of particle samples began to appear in the 1950s with improved sampling and laboratory analytical procedures for study especially of the inorganic components of particles. The theory for suspension of particles depended on the strengths of sources combined with meteorological processes dispersing the finely divided material.

After the 1950s, researchers began to suspect that chemical reactions of trace gasses or vapors in the air would produce aerosols, especially small in size. These "secondary" species that are produced in the atmosphere as condensed acids or salts generally are found in the fine particles. Inorganic species from the oxidation of gaseous sulfur and nitrogen could produce copious quantities of small particles (e.g., McMurry et al. 2004). A schematic diagram of the differentiation of particle chemistry by size is shown in Fig. 3.

Tropospheric reactions producing condensable species were identified based on laboratory experiments and field observations (e.g., Renzetti and Doyle 1959, Leighton 1961, Mohnen 1969, Cadle 1973). Went (1960) conjectured that organic vapor oxidation in the atmosphere from vegetation

hydrocarbon emissions was also a source of particles. Sulfate (SO_4^{2-}) present as ammonium salts was believed to be most prevalent secondary species along with ammonium nitrate. Went's assertion about natural aerosol production has turned out to be correct, and is enhanced by organic aerosol formation from anthropogenic hydrocarbon vapors.

Sulfur oxide chemistry to elucidate mechanisms for sulfur oxidation to form SO_4^{2-} was reviewed for possible mechanisms by Friend (1973); for gas-phase reactions by Calvert et al. (1985) and for multiple gas phase [dissolved oxygen or ozone (O_3)], aqueous and heterogeneous mechanisms including photochemistry, or metal catalysis was reported by Martin (1984). The aqueous oxidation by hydrogen peroxide (H_2O_2) was found to be most rapid during summer conditions. H_2O_2 is a product of photochemical processes. Nitrate chemistry is more complicated than SO_4^{2-} ; this component is linked with photochemistry through the product, nitric acid. Ammonium nitrate (NH_4NO_3) is found to be volatile in the atmosphere; equilibration depends on the presence of water, temperature, SO_4^{2-} , and other trace species such as sea salt. Equilibrium conditions for NH_4NO_3 favor low temperatures and minimum presence of the other acid species. Investigators calculated equilibrium conditions by accounting for the interactions of species through models such as ISORROPIA (e.g., Nenes et al. 1998).

As late as the 1980s, production of organic particles from vapor oxidation was thought to be associated with hydrocarbons of carbon number > 7 —mainly unsaturated species such as olefins or certain substituted aromatics (e.g., Grosjean and Seinfeld 1989). An example list of species

Fig. 3 Conceptual diagram of distribution by particle size with chemical species generally found in at least two modes of aerosol mass concentration. Secondary species are found in the small particle

range $< \sim 1 \mu\text{m}$ diameter and larger particles are strongly influenced by material from mechanical disintegration. (Diagram attributed to P. Mueller; from Hidy et al. 1980, courtesy of Wiley-Interscience)

identified in the organic fraction of particles determined by high-resolution mass spectrometry is given in Fig. 4.

Later, experimental results from smog chambers interpreting organics in ambient air indicated that chemical reactions of virtually any volatile organic carbon could produce condensed material (e.g., Kroll et al. 2005, Hallquist et al. 2009). Oxidation from OH and O₃ on unsaturated species was found to be catalyzed by strong acids. This finding has changed the picture of organic carbon (OC) formation in a major way and called attention not only to a range of organic species such as substituted aromatics, but also reactions of isoprene, terpenes, and other vapors, including aldehydes and amino compounds.

Recognition that the fraction of organic components in aerosols is dependent on their volatility is crucial to understanding the exchange of species between the vapor and condensed phase (e.g., Robinson et al. 2007). Pankow (1987, 1994), for example, developed an adsorption-absorption theory for partitioning of species between phases that focused on carbon number and oxidative state of particulate organics to be stable in the atmosphere. Robinson et al. (2007) noted that aerosol composition is unstable and transient in character. During “aging” in the atmosphere, particles can change their composition and size distribution significantly.

Recent investigation of the OC production in relation with interactions with acid gasses has suggested that organosulfates, organo-nitrates, or other acid related compounds are potentially important isoprene reaction pathways to form particles. This chemistry was studied in 2013 in the southeastern United States, as summarized in Carlton et al. (2018).

Most of the contemporary studies for secondary aerosol products have focused on the troposphere. Aerosol chemical reactions involving particle interactions with gasses can take place in the stratosphere as well. The Junge et al. (1961) discovery of a sulfate aerosol layer at ~20 km above ground level led to interest in a distinct set of chemical reactions for its formation. Volcanic eruptions can release sulfur compounds high into the stratosphere; sulfurous gasses are readily changed to sulfuric acid by OH oxidation. The diffusion of carbonyl sulfide (COS) from the oceans also provides another source of stratospheric SO₄²⁻ (Crutzen 1976).

An important element of stratospheric aerosol-like reactions of Cl species and NO_y species was elucidated in the 1980s that was linked with the conditions to eliminate O₃ to form the “ozone hole”. Heterogeneous reactions of hypochlorite (ClO) with ice crystals in polar stratospheric clouds were found to release Cl which reacts with O₃ (e.g., McElroy et al. 1986). The reactions of ClO and NO_y to form chlorine nitrate (ClONO₂) are suppressed in favor of depleting HNO₃ in clouds, allowing for stratospheric denitrification by ice crystal fallout or ice nucleation (Salawitch et al. 1989).

2.6 Integration via Chemical Transport Modeling

Prior to the 1970s, investigators were limited in merging aerosol microphysics with the spatial distribution of sources or the macroscale processes of the atmosphere. The continuing development of computers after the 1960s permitted their application to solve numerically fluid dynamic and chemical kinetics equations characterizing aerosol and atmospheric processes. Since the 1970s, substantial effort has been committed to development of chemical transport models (CTMs) for aerosols that integrate parameterized aerosol microphysics and chemistry with spatially resolved source emissions and meteorological processes over a range of spatial scales from urban (> 100,000) to global.

The early aerosol models of the 1980s addressed mainly SO₄²⁻ from SO₂ oxidation. The calculations were mainly regional to estimate events or annual averages. They focused on transport and mixing with rudimentary SO₂ oxidation in terms of an empirical, first-order oxidation rate. An inventory of ~40 pre-1990s CTMs is listed in Table 6.1 of Hidy (1994). Two early developments of note were the European regional model of Eliassen (1978) and ENAMAP for North America (Bhumralkar et al. 1984), both of which were aimed mainly at estimating the formation and deposition of acidic species. In the late 1980s, modelers began to assimilate more complex chemical mechanisms for SO₄²⁻ and NO₃⁻, and added a link to gas-phase reactions from photochemistry and complex oxidation reactions for SO₄²⁻ and NO₃⁻. Seigneur et al. (1999) and Seigneur and Dennis (2011) reviewed the development of CTMs, with application to air quality in North America. Two of these schemes included advanced chemical treatment were RADM and ADOM (Chang et al. 1990, ERT 1985).

Current modeling in North America relies on the US Environmental Protection Agency’s Community Multiscale Air Quality (CMAQ) model (Byun 1999) and its extension (CAMx), or the Canadian Meteorological Service model, A unified regional air quality modeling system—AURAM (McKeen et al. 2007). The codes contain parameterizations of known contributing aerometric factors to the chemistry of atmospheric aerosols. The capability of these models to simulate the atmosphere has been tested extensively with observations. These tests are summarized in the reviews of Seigneur (2001) and Seigneur and Dennis (2011). For regional or urban scale “performance,” the models can at least semiquantitatively duplicate the spatial and temporal variation in secondary species and can trace the distribution of particle from emissions. The models have assisted in interpreting very complex interactions between the microscale processes and the meteorological processes resulting in ambient concentration distributions. Such analyses obviously could not be done by simply scrutinizing concentration patterns with weather maps.

Fig. 4 Example of organic compounds found in aerosol samples at an urban site (WAW1) and a rural site in winter (DAW1) and summer (DAZ1) in Belgium. Black bar is condensed phase, white bar is vapor

phase and striped is portion volatilized during sampling. PAH are polycyclic aromatic hydrocarbons (From Van Cauwenberghe 1986; courtesy of CRC Press)

At larger spatial and temporal scales, the GeosChem model (Bey et al 2001) is currently adopted as a theoretical algorithm for many large-scale aerosol chemistry studies. The results from GeosChem calculations have been used to obtain insight about the nature of aerosols distributions across North America and other regions of the world as part of efforts to understand the role of so-called long-range intra- and intercontinental transport and chemistry on scales of 100s–1000s of kilometers.

3 Measurements

If theory is the Yin of atmospheric aerosol science, the Yang is the portfolio of measurements methods developed and ambient data collected since the turn of the twentieth century (McMurry 2002; Hidy et al. 2017; Kleinman et al. 2017). In the post-World War II years, aerosol measurement technologies have advanced rapidly in parallel with other micro-technology and remote sensing. New sampling and analytical instrumentation has enabled investigators to determine efficiently particle concentrations and composition in space and time (e.g., Solomon et al. 2014). Advances in data recording and manipulation using computers have enabled major improvements in characterization of aerosol phenomena.

Supporting the “what’s there and how did it get there” collection, measurement breakthroughs of particular importance for atmospheric aerosols include: (a) continuous determination of particle-size distributions; (b) sampling and laboratory analysis for mass concentration and composition; (c) methods for continuous determination of chemical characteristics; and (d) development of direct sensing techniques for determining optical properties of particles, and relating them to size distributions and composition.

The capability for continuous size distribution measurements is important as one of the universal characteristics of atmospheric aerosols. The identification multimodal nature of the size distributions with different environmental exposures has led to major improvements in human and ecological risk assessment. Methods important for determining chemical composition have led to source identification and to theories of aerosol chemical mechanisms. Chemical product identification has provided improvements in risk assessment of toxic chemical exposure. Advanced optical measurements has improved understanding of visibility and has led to important information about light extinction and the potential for contributing to the earth’s radiative forcing. Finally, the basic measurements of urban and non-urban aerosol properties across the troposphere and to some extent the stratosphere have given the public advanced knowledge of an important atmospheric properties related not only to phenomena of hydrometeor cloud physics and climate

change, but to environmental exposure to human and ecosystem contaminants.

3.1 Particle-Size Distributions

Because particle-size distributions have a wide range of number concentration per diameter increment, the effort to develop measurement methods has gone well beyond the counting capabilities of a light microscope ($> 0.5 \mu\text{m}$ diameter resolution). Investigators have adopted a combination of techniques that could estimate particle size and concentrations in different overlapping size ranges. The techniques used devices like diffusion batteries ($0.05\text{--}0.5 \mu\text{m}$ diameter) for sampling, inertial impaction devices ($0.5\text{--}30 \mu\text{m}$), optical scattering instruments ($0.3\text{--}10 \mu\text{m}$), condensation nuclei counters ($<0.1 \mu\text{m}$), and electrical mobility analyzers ($0.2\text{--}5 \mu\text{m}$) (e.g., Green and Lane 1957; Hidy 1984). Prior to the 1950s, these methods were rarely used simultaneously to create self-consistent number-size distributions or their particle surface or volume “moments” of the number-size distribution from < 0.1 to $> 10 \mu\text{m}$. After Junge’s (1963) effort to assemble observations into an integrated picture of number-size and volume-size distributions, a team of workers at the University of Minnesota (Whitby et al. 1972) began engineering an integrated set of instruments for routine determination of size distributions from $\sim < 0.01$ to $\sim 10 \mu\text{m}$ diameter. The methods adopted included coupling of an optical particle counter, an electrical mobility analyzer, and a condensation nuclei counter. A culmination of series of results from this system yielded a wide range of self-consistent data that better defined not only the number distribution and its components, but also verified the multimodal character of the surface and volume distributions (e.g., Whitby et al. 1972; Whitby and Sverdrup 1980). The multimodal characteristics, combined with optical observations and health studies (e.g., Lippman and Altshuler 1975), found wide acceptance for determining the key fractions, $< 2.5 \mu\text{m}$ and $< 10 \mu\text{m}$ diameter, specified in mass (volume related) ambient air quality regulations in the US and across the world (Wilson and Suh 1997).

Since before World War II, investigators established a goal of continuous measurements of atmospheric aerosols. One of the first devices that emerged during the war was the continuous nuclei counter based on a cycle of compression and expansion of humid air (e.g., Rich 1961; Skala 1963). This instrument was used initially as a combustion exhaust detector, for example, to detect submarine exhaust at sea. Later, the counter was used through the 1970s as a tool to determine atmospheric nuclei concentrations, and as noted above, was used with electrical mobility and light scattering to determine particle-size distributions. In the 1970s, opportunities for new chemical instrumentation development

provided opportunities for continuous measurement of certain aerosol species.

Commercial instrumentation has taken instruments well beyond the capabilities of the initial Whitby–Liu system. Post-1990s capabilities for particle-size determination and a number of condensation based counters now exist, so that the rudimentary approaches of the early twentieth century are far in the past for researchers interested in aerosol dynamics in size ranges. With commercial instruments readily available (e.g., www.tsi.com; www.aerodyne.com/products/), both researchers and workers interested in monitoring have easy access to size-differentiated measurements over a range from $< 0.002\ \mu\text{m}$ to $> 10\ \mu\text{m}$ diameter. These capabilities offer major opportunities for measuring aerosol physical properties extended in space and time.

3.2 Mass Concentration and Particle Composition

The Junge and Whitby–Liu observations stimulated a strong interest in improving measurements of aerosol mass concentration and composition as a function of particle size. Through the 1970s, measurements of particle chemical composition were carried out by sampling on a substrate, including filter media, followed by laboratory analysis for various species (e.g., Lodge 1988; Willeke and Baron 1993; Spurny 1999). Sampling methods evolved with size differentiation using both impaction and diffusional deposition, and filtration. Impactors, for example, were extended to a low operating pressure regime to obtain particle both by inertial processes and diffusional deposition. Filtration was improved for well-defined size fraction (e.g., Watson et al. 1989), and advanced knowledge of filter substrate adsorptive performance, particle volatility, or chemical reactivity with trace gas volatility (e.g., Appel et al. 1980; Chow 1995; Chow and Watson 1998).

Because of the regulatory driving force in the 1970s, strong emphasis was placed on mass determination in certain particle-size ranges associated with human health effects inferred from epidemiology and respiratory exposure studies in the laboratory (e.g., Lippman and Altshuler 1975) and visibility impairment (e.g., Chow 1995; Watson 2002). Particle mass concentration for many years was obtained with a standard 24 h collection from so-called high-volume samplers, which amounted a vacuum cleaner pump hooked to a filter holder (e.g., NASN 1965; Watson et al. 1989). Samples were then transferred to a laboratory for gravimetric measurement of filtrated mass.

In the 1970s filters evolved to include multivolume devices, including the virtual impactor (e.g., Marple and Olson 2011) and the automated sequential sampler. These samplers enabled improvements in the quality of gravimetry, but also in variable sampling time, including diurnal resolution. Such methods enabled researcher investigations

of diurnal chemical characterization that greatly improved knowledge about the variability of particle concentrations, and the nature of particle chemistry paralleling gas-phase chemistry (e.g., Whitby et al. 1972; Hidy et al. 1980; Wolff et al. 1981).

A portfolio of automated laboratory analytical tools having low detection and quantification limits. These methods enabled routine observations of bulk composition to take place (e.g., Fehsenfeld et al. 2004). With routine chemistry, the major “universal particle” components were identified, including sulfate and nitrate as ammonium salts, crustal species, sea salt and black and organic carbon. Determinations of bulk chemical constituents by a variety of methods, including atomic absorption spectroscopy, ion chromatography (Chow and Watson 1998), automatic colorimetry, X-ray fluorescence (XRF), proton X-ray-emission analysis (PIXE), inductively-coupled plasma mass spectroscopy (ICP-MS), or thermally differentiated carbon (e.g., Chow and Watson 1998), enabled investigators to obtain “closure” for particle mass balances. With the formalism of analytical chemistry, particle data were “certified” for quantitative quality using reference methods of quality control and assurance applying standard reference methods (e.g., EPA 1994, 2013; WMO 2001). These methods were complemented with single particle measurements using microscope capabilities, mainly electron microscopy (Casuccio et al. 1983).

With routine particle chemistry emerging in the literature, advanced studies adopted special laboratory measurements of particle chemistry that began to explore hypotheses about particle components (Chow et al. 2018). These included use of neutron activation analysis for light elements (Ragaini 1980), and exploration of valence state of sulfur and nitrogen using photoelectron spectroscopy (Novakov et al. 1972).

Instruments for continuous measurements of mass concentration began to appear in the 1970s with beta radiation absorption gages (Wedding and Weingard 1993), beta attenuation monitors (BAM) and the Tapered Element Oscillating Microbalance—TEOM (Lewis 1981). Both BAMs and TEOMs are currently used in many US and international air monitoring programs to complement filter-gravimetry for mass determinations. The TEOM instrument depends on tracking the change in oscillation of a glass tube with loading of aerosols from the atmosphere. Instruments for measuring continuously particulate SO_4^{2-} were reported using modified SO_2 gas analyzers (e.g., Mueller and Collins 1980; Hering 2005), later the versatile particle in liquid sampler for water soluble anions and cations (PILS) (Weber et al. 2001; Orsini et al. 2003), and the aerosol mass spectrometers (AMS) (Canagaratna et al. 2007), Aerosol time of-flight mass spectrometry (ATOFMS) (Suess and Prather 1999) or ionization and photon ionization mass spectrometry (Hanley and Zimmernan 2009).

Intercomparisons of different SO_4^{2-} detectors were held in 2001 (e.g., Drewnick et al. 2003). Intercomparisons for NO_3^- measurements were reported by Stolzenberg and Herzig (2003). Semicontinuous thermal differentiated measurements of black carbon and organic carbon derived from research of several groups including Huntzicker et al. (1982), Turpin and Huntzicker (1991) and Chow et al. (1993). Sometimes applied as a complementary black carbon method, a light absorbing carbon measure uses the aethelometer (e.g., Hansen et al. 1982; Arnott et al. 2005). Carbon analyzers are now used routinely in monitoring (e.g., field samples to laboratory, Malm et al. 1994 and field measurements, Hansen et al. 2003; Solomon and Sioutas 2008). Recent advances from single to multiple wavelength carbon analysis allows the separation of black from brown carbon (e.g., mostly in the smoldering phase of biomass combustion and secondary organic aerosol) that absorb light at lower wavelength (Chow et al. 2018). Empirical, multiple carbon species classification by “source” as hydrocarbon-like or oxidized species now uses the AMS instrument for semicontinuous observations (Jimenez et al. 2003; Zhang et al. 2005).

The potential for major improvements in particle measurements by established collection methods and laboratory analyses, and the new continuous instruments were demonstrated in a US study called the “Supersites” program (e.g., Solomon and Sioutas 2008). This project involved seven sites located across the US that were instrumented with conventional and new particle and gas instrumentation to characterize particle chemistry and their origins in the 2.5 μm diameter range and larger in support of a new, mass-based US ambient air quality standard. The study revealed major improvements in knowledge of aerosol properties, and demonstrated the strengths and weaknesses of instrumentation for continuous observations.

The Supersites initiative was complemented by a multi-year program in the southeastern US called the SEARCH (Southeastern Aerosol Research and Characterization) study (e.g., Hansen et al. 2003). From 1999 to 2016, the SEARCH undertook a program comparing urban and rural aerosols and gasses at eight sites using basic measurements accompanied by special studies using advanced research instruments and techniques (e.g., Zheng et al. 2002; Carlton et al. 2018).

One of the more dramatic 20th Century achievements in characterizing atmospheric aerosol chemistry addresses the organic fraction (e.g., Mader et al. 1952; Jacobson et al. 2000). The presence of organic species in aerosols was recognized some time ago, but most of the effort to specify this fraction relied on bulk solvent soluble extracts (e.g., NASN 1965). In the 1970s, high-resolution mass spectroscopy and other contemporary analytical instruments were applied to speciate organics (e.g., Schuetzle et al. 1973; Simoneit 1986; Van Cauwenbergh 1986; Rogge et al. 1993). Additional speciation capabilities are facilitated with solvent extraction

followed by gas chromatography–mass spectroscopy (Zheng et al. 2002). Investigators found a very large number of organic compounds in atmospheric samples that ranged from alkanes and poly-aromatics to dicarboxylic acids. Speciation methods provided for identification of only a fraction of species present in aerosols, making an organic mass balance problematic. An example of analysis by molecular categories is shown in Fig. 4.

Making the organics characterization complicated is the fact that these species are derived from direct source emissions, including incomplete combustion of fuels, to material produced in the atmosphere from oxidation and other reactions of organic vapors (e.g., Grosjean and Seinfeld 1989; Hallquist et al. 2009). Furthermore, the organic fraction exhibits volatility that creates conditions for exchange. The volatility of aerosol organics is exemplified by data in Fig. 4 for different groups of species as a function of molecular weight or carbon number. Dealing with volatility in terms of partitioning species by phase was examined in detail in Pankow’s (1987, 1994) work.

The complexity and uncertainty in the organic species directed aerosol scientists to seek a simplified approach to account for the organic aerosol component. The two methods described above are thermal differentiation with carbon detection and AMS with separation by hydrocarbon-like and oxidized-like speciation. Speciation of organics in the atmospheric aerosol has usually accounted from about 20% or less of the total organic fraction. The simplified methods have allowed for “closure” of the aerosol mass balance with certain assumptions about the relation between organic matter and organic carbon (Turpin and Lim 2001; El-Zanan et al. 2009; Chow et al. 2018; Riggio 2018). However, the detailed mechanisms for formation of organic components vs. derivation from sources still require knowledge of organic speciation in the particles.

Interaction between organic vapors and acids in the atmosphere facilitates formation of secondary organic carbon in particles (Jimenez et al. 2009; Hallquist et al. 2009). A major investigation of particle chemistry including the interactions of organic vapors with acid gasses took place in U.S. Environmental Protection Agency 2013 at sites in the southeastern US (Surratt et al. 2008; Carlton et al. 2018). This campaign involved early summer experiments with an array of continuous particle instruments at a rural ground site and a tower for observations aloft, with overflights from aircraft. The results reported in Carlton et al. (2018) support the theoretical pathways for aerosol production from acid gas interactions with natural and anthropogenic hydrocarbons in a photochemically active environment. The project extended major chemistry knowledge of secondary aerosol carbon production described in Hallquist’s (2009) review.

A long-standing ambiguity in characterizing aerosol particles is their interaction with water vapor. When humidity

increases particles absorb water because of their hygroscopicity even in conditions well below supersaturation. Vapor–liquid equilibrium considerations (Kohler 1936; Winkler and Junge 1972; Hanel 1976) specify an expected water absorption depending on particle size and water vapor pressure. Aerosols play a key role in hydrometeor formation and growth (e.g., Pruppacher and Klett 1978). Moreover, the presence of water in particles is an important factor in determining mass concentration from bulk composition, as well as chemical processing. Measurement of water content has been attempted through various means from examining changes in collected particles on substrates to optical devices with varying humidity (e.g., Winkler and Junge 1972, Covert et al. 1980). Direct measures in the atmosphere derive from microwave determinations (e.g., Ho et al. 1974). Variations with composition and temperature have been reported by Tang and Munkelwitz (1993), Cruz and Pandis (2002) and Kreidenweis et al. (2005). Most of the attention to hygroscopicity focused on inorganic species; however, Saxena and Hildemann (1997) reviewed this phenomenon for organic species.

Improvements in sampling technology have enabled not only ground-level particle collection, but have provided the basis for sampling aloft from the troposphere to the stratosphere. Methods aloft had to account for a principle of isokinetic sampling aboard a balloons or aircraft, along with pressure–volume variations with altitude. Sampling the troposphere and stratosphere began with Junge’s work (e.g., Junge 1955; Junge et al. 1961; Junge 1963), and a long-term mid-continental record of size distributions (Deshler et al. 2003). Aircraft sampling in the troposphere has been reported by Blifford and Ringer (1969), Hobbs (1993) and Clarke and Kapustin (2002) to obtain knowledge of aerosols over the continents. Notable in this period was the use of instrumented commercial aircraft to sample aerosols in the upper troposphere and lower stratosphere (e.g., Hogan and Mohnen 1979; Hermann et al. 2007). A variety of light aircraft sampling was conducted as support for urban and regional experiments (e.g., Blumenthal 2011; Blumenthal et al. 1981; Collins et al. 2000) to global sensing over remote locations. Some major experiments to characterize tropospheric aerosols in the 2000s over the Pacific Ocean in the northern and southern hemispheres (e.g., Clarke and Kapustin 2002). Measurements of dust from Africa across the Atlantic Ocean also took place in the 1990s (e.g., Prospero 1996) and pollution over India and the Indian Ocean was studied (Lelieveld et al. 2001). These campaigns greatly improved the knowledge of hemispheric aerosol conditions, including documentation of the occurrence of intra and intercontinental aerosol transport.

3.3 Optical Sensing

Light extinction by airborne particles complements the measurement of mass concentration and composition. Monitoring of light extinction has taken place well before the 1950s using a surrogate measure of visual range from airport records (e.g., Middleton 1952). Contemporary aspects of visibility impairment by air pollution haze have been investigated by Trijonis (1990) and Schichtel et al. (2001) and have been a major issue addressed for conditions in the pristine areas of the US (e.g., Malm et al. 1994). As an alternative to visual observations, light extinction has been inferred in terms of light scattering and light absorption coefficients determined at a point with nephelometers or photometers (Watson 2002). These observations oversimplify the complexities of visibility in terms of the interpretation of light contrast and human perception of “seeing” objects through a viewing distance, as discussed by Middleton (1952), and Henry (1987, 2002), for example. Quite apart from visibility questions, light extinction and optical depth are used routinely to document vertical distributions of aerosol optical properties. The importance of aerosol to atmospheric turbidity and consequent radiative influence has been studied extensively since the 1970s (e.g., Rasool and Schneider 1971, Toon and Pollack 1976, Toon et al. 1988, Twomey 1991, Charlson et al. 1991).

Long-term light scattering and absorption values have been measured or calculated from aerosol mass and composition in conjunction with the US Interagency Monitoring of Protected Visual Environment (IMPROVE) program (e.g., Malm et al. 1994). Urban–rural comparisons have been reported for sites in North America by Delene and Ogren (2002) and in the southeastern US (Hansen et al. 2003).

Open path measurements of light extinction using passive devices such as sun photometers (Shaw, 1982; MacArthur et al., 2003) or active devices such laser photometers or lidar (Kreid 1976; Uthe 1983) have been applied in the 1970s as a replacement for visual range; observations have served to estimate aerosol light extinction or aerosol optical depth (AOD) in the atmosphere. Optical depth has been observed extensively with upward looking instruments and downward viewing from satellites since the 1970s (e.g., King et al. 1999, Hoff and Christopher 2009).

Aerosol optical properties and the Earth’s radiative conditions are of great interest for extending knowledge about the Earth’s radiative balance (Chandrasekhar 1960) and climate change (e.g., Charlson et al. 1992, Penner et al. 1995). Investigators recognized the significance of aerosol effects on radiation well before the wave of climate modeling issues emerged after the 1980s, with studies of energy scattering (e.g., Toon and Pollack 1976; Hansen et al. 1980), and absorption (Bond et al. 2013). Black and brown carbon have become increasingly important for observations given

their contribution to absorption of light and broad wavelength radiation (Andreae and Gelencsier 2006; Bond et al. 2013). Long-term measurements needed to document aerosol forcing of radiative changes have employed major efforts, including AERONET, a global ground level surveillance of aerosol optical properties by remote sensing from the ground (e.g., <https://aeronet.gsfc.nasa.gov/>), and by AOD measurements from satellites (e.g., Hoff and Christopher 2009).

3.4 Source Identification and Quantification from Measurements

Much of the late twentieth century interest in atmospheric aerosols involved the sources of airborne particles. Substantial effort has been devoted to inventorying natural and anthropogenic sources for particles by mass and composition (e.g., Miller et al. 2006; Streets et al. 2003). Complementing the integration of theory in source-based models is so-called “receptor-based” analogs to identify sources of atmospheric aerosols in terms of their ambient composition. These methods take advantage of the extensive and sustained observations of the bulk or speciated chemistry of aerosols (e.g., Friedlander 1973; Watson 1979; Hopke 1985; Schauer et al. 1996; Henry 2003; Shrivastava et al. 2007). The approach is a straight forward that assumes that the particle composition from sources (e.g., lead halide for pre-1980 gasoline-powered vehicles, silicon–calcium for soil dust or sodium chloride for sea salt) essentially can be found as a fraction of species in ambient particles [a chemical mass balance (CMB) or conceptual derivative thereof]. These methods have been used extensively to identify sources or source categories represented by aerosol chemistry. Until recently, the methods were unable to differentiate between sources

of secondary species, but if sufficiently detailed, simultaneous gas-and particle-composition data are available, this differentiation can be achieved (e.g., Blanchard et al. 2012).

An example of source apportionment for an urban winter aerosol is given in Fig. 5. This identification shows that motor vehicles and sulfate–nitrate–ammonium account for most of sources in this case. The differentiation of transportation and other sources depends on “source profiles” representative of real-world emissions, which drive the apportionment. Currently used solutions to the CMB equations (Watson et al. 2016) include effective variance least squares, positive matrix factorization (PMF), and edge discrimination rely on the knowledge of the source profiling, and use them indirectly to interpret results of apportionment.

The CMB approach has also been used to apportion particle light extinction to sources (e.g., Wolff et al. 1981; Watson et al. 1998). This method assumes a linear summation of chemical contribution by source to overall light extinction, with the composition or source contributions weighted by scattering or absorption efficiency of species (Watson et al. 2002; Malm et al. 1994; Pitchford et al. 2007). Light scattering involves all particle sources, both primary and secondary. Absorption is mainly associated with black or brown carbon, though a small contribution can come from iron particles.

4 Highlight and Highlighters’ Hierarchy

The past 100 years has seen the advancement of atmospheric aerosol science on both theoretical and observational grounds following the fundamental physics of the 19th and early twentieth centuries. Foundations for atmospheric aerosol science are derived from early knowledge of atmospheric phenomena (e.g., Husar 2000; Podzimek

Fig. 5 Example of source apportionment from bulk chemical composition of fine particles determined from measurements in the Denver, Colorado metropolitan area in winter 1997–1998. Note the fractions of particles as $(\text{NH}_4)_2\text{SO}_4$, NH_4NO_3 and the carbon containing fractions. (From Watson et al. 1998)

Fig. 6 Illustration of five generations of highlighters who have contributed substantially to the progress in atmospheric aerosol science. The identified generations are separated arbitrarily for their work mostly in theoretical developments vs. mostly in [measurements](#). Most of the researchers cited have been involved in both of these elements of the atmospheric science. The investigators are illustrative but not inclusive; Topics associated with highlighter contributions are listed in the Supplemental table (Online Resource 1)

2000). The fundamentals enabled aerosol science to advance from the wisdom of many scientists, including Einstein for Brownian diffusion, Smoluchowski for coagulation, Becker-Doring for nucleation, Wilson for

nucleation and the optical physics of, Rayleigh, Mie, and Tyndall. The syntheses of Whytlaw Gray and Patterson, Green and Lane and Fuchs set the stage for aerosol physics to develop in the post-World War years of the twentieth

century. By the 1950s, the cast of characters expanded with scientists and engineers motivated not only by persistent technical questions about airborne particles, but also practical concerns for environmental protection and climate change.

Looking at the investigators mentioned here to exemplify highlights in aerosol science, one could arbitrarily assign the nineteenth century and early twentieth century group in archeologists jargon to an “archaic” period. After this group, there are at least six generations of investigators who have contributed to current knowledge of atmospheric aerosols. A first generation that represents contemporary atmospheric aerosol science emerged after the 1940s, as illustrated in Fig. 6. From this group, successive generations emerged as aerosol science has advanced perhaps exponentially in building a foundation of experience on both theoretical and experimental-observational grounds. Five generations that are highlighted are shown in Fig. 6. The groupings are indicated subjectively in terms of mostly theorizing vs. measuring. Individuals indicated are for illustration and for implying a connection between mentors in the previous generations and the “succession” of investigators following up on both theoretical and measurement issues suggested in examples from the references.

These choices in Fig. 6 represent only a portion of the many people studying contemporary atmospheric science. In the scheme, some of major contributors are listed exemplifying highlighted progress in the designed important areas of knowledge since the 1950s. The sixth generation of students and recent graduates has not been identified—time will tell with which of these investigators will come forth with future contributions to exemplary aerosol science.

Most of those highlighted in Fig. 6 have not only contributed to atmospheric aerosol science, but also through their laboratories and institutions have fostered and mentored the succeeding generations. The research developed over the past 70 years has greatly expanded our understanding of atmospheric aerosols, not only “what’s there and where do the particles come from,” but the consequences of their presence as catalysts for hydrometeor clouds, environmental risk, and through changes in radiative forcing.

Looking to the future of those “in practice”, one can only guess what the next steps in science will be. Such progress will be dictated in part by the interests and concerns of the public and their representatives, who will supply investigators’ resources. Continued effort will address improvements in aerosol processes important to estimating concentration distributions, and more importantly interactions between particles and clouds that are crucial for modeling weather and climate-related processes.

It would seem that there will be emphasis moving towards recording changes with sources and chemistry in ambient aerosol conditions not only at the ground but aloft. Emphasis

is likely to continue on the characterization of the organic particle fraction, since its complexity in species origins and potential toxicity remains to be elucidated. As a part of this direction, emphasis is likely to be placed on the biological component of organic species. Although this portion of the fine aerosol by mass is small, its implications for human and ecosystem risk are suspected to be inordinately high relative to inorganic material. The Frohlich-Nowoisky et al. (2016) review of current knowledge bioaerosols offers an initial foundation for extended research of this suspended material; its complexities perhaps more than equal to the current picture of other organic aerosols.

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Compliance with ethical standards

Conflict of Interest No conflict of interest for the author of this manuscript.

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