Developments in Atmospheric Science, 7

Atmospheric Aerosols

by

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Foreword

The atmospheric aerosol affects our weather, our seeing and in some instances our well-being and even survival. It is now receiving a great deal of attention from chemists, engineers, physicists and meteorologists. My intention in writing this text has not been to attempt an encyclopaedia of the aerosol, nor has it been to produce a definitive text on the subject or a portion thereof: rather has the aim been to try to tell the story of the atmospheric aerosol in (hopefully) a reasonably coherent way — to put together parts of the subject which are treated in greater depth, but in a more isolated context, in books such as Füchs "Mechanics of Aerosols", Hidy and Brock's "Aerodynamics of Particles", van de Hulst's "Light Scattering by Small Particles", and so on.

Involving as it does virtually all classical physical disciplines as well as climatology, meteorology, atmospheric optics, radiative transfer and several branches of chemistry, the subject is far flung and at times ill-defined. When writing about it one is continually invited to digress. In attempting to produce a reasonably coherent text I have found it necessary to be at times arbitrary; some readers will be greatly dissatisfied by the omission of any discussion of the now very numerous chemical analyses of aerosol material which has been published, of photochemical smog, of the gaseous reactions leading up to gas-to-particle conversion, of the deposition of aerosol particles in the lungs and respiratory tracts, and many other subjects which are related to the atmospheric aerosol and are of undisputed importance. For these omissions I can only offer my apologies.

A great many people helped in the preparation of this book. I acknowledge with gratitude use of the facilities of the C.S.I.R.O. Division of Cloud Physics which were placed at my disposal by Jack Warner. Barbara Georgeson and Joyce Barnett made valuable contributions to the preparation of the book. Over the years my own work in the field has benefited from a great many people. The wisdom and experience of the late Leo Wenzel Pollak and the dedication and great technical ingenuity of Ken Davidson have been of especial value. I am grateful to the Elsevier Scientific Publishing Company for their invitation to write this book and their willing acceptance of delays and postponements.

S. TWOMEY

List of Frequency Used Symbols

A	Hamaker constant for non-retarded forces
B	mobility; Hamaker constant (retarded forces)
C_D	drag coefficient
D	diffusion coefficient
E	electric field; collection or deposition efficiency
F	flux
G	multiplicative factor in drop (particle) growth equation
H	magnetic field
I	intensity
J	Bessel function of the first kind
K	eddy diffusivity; dielectric constant
L	latent heat
M	molecular weight
M_W	molecular weight for water
M_A	molecular weight for air
M_N	molecular weight of nucleus
N	total or cumulative concentration
N_A	Avogadro's Number
Pe	Peclet Number
Q_a	efficiency factor (absorption)
Q_e	efficiency factor (extinction)
Q_s	efficiency factor (scattering)
R	universal gas constant per mole
Re	Reynolds Number
S	supersaturation
Stk	Stokes Number
W	liquid water content
a	particle radius
e	vapour pressure
g	free energy per molecule (chemical potential); acceleration of gravity; asymmetry factor
h	spherical Hankel function
i	van 't Hoff factor; $\sqrt{-1}$
i_1, i_2	Mie intensity functions for scattering of light by a sphere
j	spherical Bessel function
k	Boltzmann's constant; extinction coefficient
k_e	extinction coefficient (total)
k_s	extinction coefficient for scattering
k_a	extinction coefficient for absorption
m	refractive index
$\hat{n}$	normal unit vector
p	pressure; scattering phase function

q	electric charge
r	particle radius; length of radius vector
u	volume; velocity
u^*	friction velocity
$\overline{u}$	screen velocity
v, V	volume
x	size parameter $2\pi r/\lambda$
z	height
α, β	power of radius in power distributions
γ	ratio of specific heat
ϵ	mass transport rate through a size spectrum
η	kinematic viscosity
θ	constant angle; temperature; scattering angle
κ	thermal conductivity
λ	wavelength; mean free path
μ	cosine of direction angle
ν	size distribution; dynamic viscosity; kinetic coefficient $\sqrt{RT/2\pi M}$
ρ	density
ρ_a	density of air
ρ_p	density of particle
σ	surface free energy; stress
τ	optical depth or thickness
ϕ	azimuth angle
ω	angular frequency
$\overline{\omega}_0$	single scattering albedo
ΔG	Gibbs free energy increment
ΔG^*	critical value of ΔG

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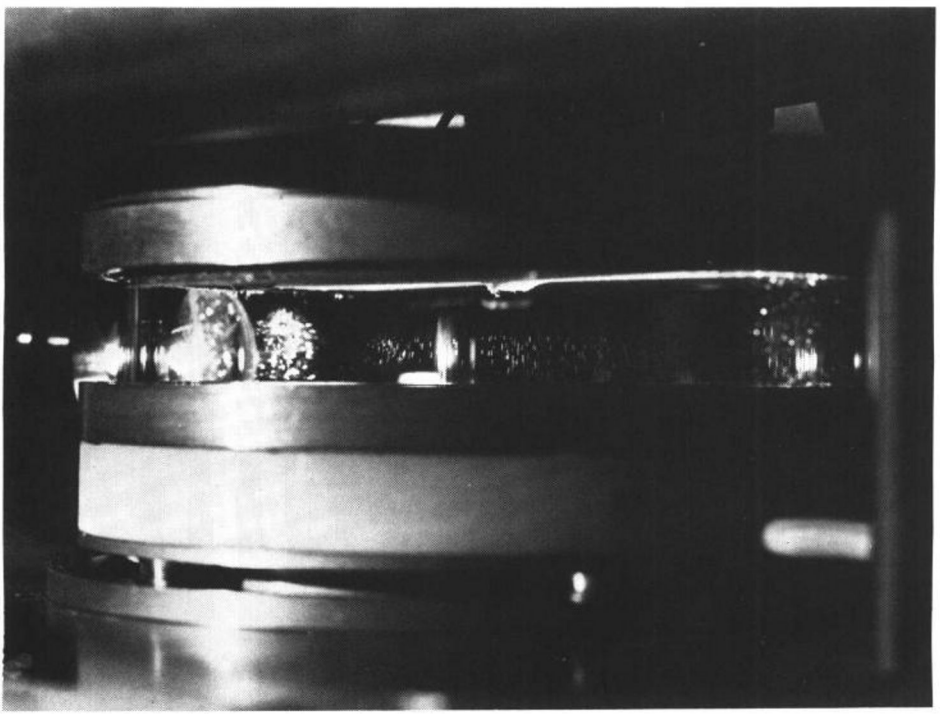

Droplets (around 2 μm in radius) growing in a thermal diffusion chamber.

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CHAPTER 1

Introductory Topics

With a generally increasing awareness of the finite size of our earth and atmosphere, all of us are becoming more concerned with all kinds of pollution and are less likely to fob off problems of pollution on the "almost infinite" size of the atmosphere and ocean — which for many years has tacitly or vocally been invoked as a safe and dependable buffer which could absorb without adverse repercussions any effluents which mankind might see fit to disgorge into it. This has led to an increasing awareness and a greater interest among individuals, society and governments in the whole question of atmospheric (and oceanic) composition and man's effects on it. The sources and sinks of atmospheric particles and the properties, lifetime and changes undergone by airborne particulate material are of especial interest, firstly because particles are more readily noticed than the mainly invisible gaseous pollutants and secondly because particles are a link in the chain of the removal process which returns gaseous pollutants to the earth's surface. This point hardly needs elaboration, but it is sometimes overlooked that it is not alone the *rate* of emission of a possibly noxious pollutant that matters, it is the product of rate of emission and "residence time" (average time before return to the surface by diffusion, fallout, rainout or other similar mechanisms). If, for example, a rate of emission of 100 megatons per year is considered — and quite a few natural and man-made emissions occur at this rate — then if the residence time of the pollutant is 10 years, some 10^9 tons will reside in the atmosphere in a balanced steady-state situation, which will give an average concentration of about 10^{-7} g/litre or 100 parts per billion (ppb), since the mass of the atmosphere is about 10^{22} g or 10^{10} megatons. If on the other hand the residence time is only a few hours, the steady-state concentration is proportionally lower, around 10^{-11} g/l or 0.01 ppb. In the case of many atmospheric pollutants either particles or cloud and rain drops or both are closely involved in the removal process, which may run gas → particle → cloud droplet → rain drop → surface deposition or, alternately, gas → cloud droplet → rain drop → surface. In either event dry particles are involved (a nucleus is required for each cloud droplet) and a gamut of processes of particle formation and growth, and cloud droplet formation and growth, are crucial. An enormous range of dimensions is spanned by these processes and it is important to note that a process which is dominant in one size range — or an approximation that is good in one size range — may be quite irrelevant at much larger or much smaller sizes. The following paragraphs will briefly

outline the dimensions involved. We will primarily use c.g.s. units but will deviate from these where it seems appropriate — for example, units of megatons per year or tons per day are more easily grasped in an order-of-magnitude sense than grams per second. The particle radius unit, the micron (μ or μm) = 10^{-4} cm = 10^{-6} m = 10^{4} Å, is especially common and useful, especially since a radius of 1 μm approximately separates particles which are individually visible and measurable (e.g., in an optical microscope), which fall at appreciable rates and which undergo very little Brownian movement, from smaller particles which can be seen only ultramicroscopically but not resolved, which fall at a negligible speed and which are dominated (so far as their main properties are concerned) by Brownian movement.

1.1. DIMENSIONS

The dimension of a small molecule or an atom is of the order of 10^{-8} cm. There is no generally accepted definition of how large clusters of molecules must become to be particles, but particles with radius about 10^{-7} *cm* have been measured and according to some workers this represents the smallest particle size detectable by condensation nuclei counters (which are a convenient and commonly used way of measuring particle concentrations in air samples). Particles of this small size undergo large Brownian movements which allow them to reach the walls of any reasonably sized container in a few seconds or at best a few minutes, and they are not readily sampled and undergo large storage losses; particles of this size also coagulate very quickly with larger particles, for the same reason. It is worth commenting at this point that all or most particles which reach the walls of a container are believed to stay there, and there is a body of experimental evidence (albeit indirect) that adhesive forces are strong enough to ensure this; molecules, on the other hand, do not behave in this way and one could envisage the distinction between molecules and particles being based on adhesion to clean walls but our knowledge of adhesive forces is not very good and so far as this writer is aware distinction on these grounds has not been attempted.

10^{-6} *cm* represents a more stable and permanent size, at which reasonable storage times are possible and coagulation at atmospheric concentrations is not excessively fast. Direct observation of particles by electron microscopy becomes possible for particles around this size or larger.

10^{-5} *cm* is considered to be "large" in the context of the atmospheric aerosol. It also is a size region which is little affected by both Brownian movement and by gravitational settling and particles of this size are likely to have the longest survival as individuals since both diffusive and inertial coagulative processes are inefficient. 10^{-5} cm also represents a size of particle which is very difficult to produce directly: it is virtually impossible to grind solid material to this degree of fineness while condensation from the

vapour phase (except with very volatile materials) tends to give smaller particles.

10^{-4} cm (1 μm). In the parlance of atmospheric aerosol workers this is the small end of the "giant" particles in the atmosphere. The falling speed under gravity of a 1-μm particle is only about 1 mm every 5 seconds, but even this slight sedimentation amounts to 20 m per day — and falling speed increases quadratically with radius in this size region. These particles can readily be observed on a surface under moderate magnification, but they cannot be sized very well.

10^{-3} cm (10 μm). This is the approximate size of cloud droplets, which of course are a very important and special subgroup of the atmospheric aerosol. The falling speed of a 10-μm particle of density 2 g cm^{-3} under normal surface conditions is about 2 cm sec^{-1}, so in a couple of minutes most 10-μm particles in an average room would fall out under gravity. Particles of this size lying on a suitable surface can be seen with the naked eye and their dimensions can be measured under the (optical) microscope.

10^{-2} cm (100 μm). This is the size of fine drizzle drops (falling speed about 1 m sec^{-1}). In fine weather everyday experience shows that particles of this size are absent — or at least extremely rare — except in dust storms and other rare violent episodes, natural or caused by man. Drops of this size are produced by sea spray, but fall out quickly and do not usually travel very far from their source.

10^{-1} cm (1 mm). A typical size for raindrops. Something like 4×10^{22} raindrops per year are produced by the atmosphere, or about 10,000 per square centimetre of surface, but the volume concentration is slight — about 10^{-5} cm^{-3} (10 per cubic metre) in moderate rain, while the averaged lower atmosphere raindrop concentration (not a very meaningful number) is only about one hundredth of the latter figure.

1 cm. Falling raindrops break up through hydrodynamic effects around 0.5-cm diameter and liquid drops of 1-cm radius are not found. However hail, graupel and snow ("solid hydrometeors") can attain this size.

10 cm. Occasionally hail particles of this size are reported. Severe damage is caused, of course, by such hailstones.

>10 cm. One is now in the region of very rare occurrence indeed. For practical purposes one may say that 10 cm is the upper limit of atmospheric particles. Of course aircraft, meteorites and volcanic fragments will be larger.

The above brief categorization extended from 10^{-8} cm to 10 cm, and even if we excluded the rarer or more transient particles there still remains an enormous range, more than six decades from 10^{-7} cm to a few millimetres. In terms of mass or volume this encompasses about twenty decades and the range of concentration values is almost as great. It is important to realize quite clearly that the range from 1 μm down to molecular size extends over as many decades as from 1 μm up to large hailstone sizes and that bulk descriptions such as "aerosol number concentration", "mean size" and so on must be defined and used with great care.

1.2. PARTICLE SIZE DISTRIBUTIONS

1.2.1. General aspects of size distribution functions and their use

Solid aerosol particles impacted on slides, caught on spider threads or precipitated in thermal or electrostatic precipitators are rarely seen to be spherical — at least so far as those particles are concerned which can be seen in the microscope or electron microscope. As one approaches molecular dimensions it is unlikely that spherical groupings of molecules will often be preferred. Even though spherical particles are likely to be rare, most discussions of particle size in the field of aerosol physics tend to treat the particles as spherical and give their size in terms of a single dimension usually quoted as "radius" or "diameter".

Microscopically visible particles also differ in colour, texture, uniformity and in water-solubility and adsorption. Unfortunately, if we attempted to categorize the aerosol content of 1 cm^3 of urban air, particle by particle, including detailed descriptions of shape, structure, etc., for each particle as well as similar details for any smaller particles which might be found attached to larger ones, the task would consume a lifetime and would be quite worthless, since long before this the contents of the cubic centimetre would have changed many times as a result of evaporation, condensation, coagulation, adsorption, desorption, as well as chemical processes in bulk and at surfaces. We are therefore forced to lower our standards and to resort to drastically simpler descriptions in which particles are grouped according to a single size parameter. The precise meaning of the size is often vague; in some instances mass or volume may be determined and the size of the particle quoted as the equivalent spherical radius (i.e., the radius of a sphere of equal volume); in other instances the diffusion coefficient of the particle is the fundamental quantity determining the separation by size and the particle is in effect equated to a sphere of the same diffusion coefficient.

To furnish even the roughest reasonably comprehensive description of the atmospheric aerosol we must cover the range from the smallest particles with appreciable lifetimes, perhaps about 10^{-7} cm in radius, to the largest particles which may be expected to turn up occasionally in perhaps tens or hundreds of cubic metres of air, which means certainly 10 μm (10^{-3} cm) and perhaps 100 μm. If we admit cloud, fog, rain and snow as part of the aerosol (which they certainly are, unless one is thinking only of the so-called "dry aerosol") then the upper limit must be set at around 10 cm (large hail). The radius range 10^{-7} to 10^{-4} cm encompasses nine decades in mass, the range 10^{-7} to 10 cm some twenty-four, and no single technique can be expected to be useful over such a range. We are therefore forced to patch together measurements obtained by widely different techniques in order to cover the wide range of sizes involved. If one technique measures mass and another, say, electrical mobility, there is no one-to-one relationship between

them (electrical mobility is independent of density) even for a constant shape of the particle. However, the range of variation of densities of known substances is not large (perhaps from 0.7 to 4 g cm^{-3} for commoner solids) and the artifice of relating both measurements to a particle radius is not misleading unless carried too far. It is notwithstanding an artifice.

Having agreed to describe the aerosol in terms of how many particles have radii (in the loose sense just described) in a given interval, there are still a number of choices open. The most obvious description is that given by the direct "*radius distribution*" $\nu(r)$, which implies that there are $\nu(r)\Delta r$ particles with radius between r and $r + \Delta r$. This function is conceptually simple and direct, but it has practical drawbacks. There may be in a given sample, say, 5000 particles cm^{-3} less than 10^{-6} cm, 5000 particles between 10^{-6} and 10^{-5} cm and perhaps 1 particle cm^{-3} greater than 0.1 μm. If we attempt to plot this on a linear scale, with, say, $x = 20$ cm corresponding to $r = 5$ μm, we find the entire region $r < 2.5 \times 10^{-6}$ cm (where almost all the particles are found) to be cramped into the first millimetre, and the distribution when plotted is indistinguishable from a single huge spike at the origin. To open out the horizontal scale the value of a logarithmic scale is obvious, and when dealing with magnitudes extending over several orders of magnitude it is natural to think logarithmically as well as plotting logarithmically, and the use of $\log r = x$ as a size measure is both natural and convenient. This leads to the *log-radius distribution* $n(x) = n(\log r)$ such that there are $n(\log r)\Delta \log r$ particles with log radii between $\log r$ and $\log r + \Delta \log r$. [A slight drawback to this usage is the existence of decimal and Naperian logarithms, but this is not serious. Naperian logarithms are algebraically simpler and we choose to use them here. To change from one to the other one only needs to note that if n' refers to the value when decimal logarithms are used, then:

$$\Delta n = \Delta n' = n(\log_e r)\,\Delta(\log_e r) = n'(\log_{10} r)(\log_{10} r)$$

The log radius distribution functions are therefore related through $n'(\log_{10} r)/n(\log_e r) = \Delta(\log_e r)/\Delta(\log_{10} r) = \log_e 10 = 2.3026$.]

The obvious and "natural" distribution function $\nu(r)$, such that there are per unit volume of air $\nu(r)\Delta r$ particles with radius between r and $r + \Delta r$, is of course readily found from $n(x)$, since evidently $n(x)\Delta x$ must equal $\nu(r)\Delta r$ when $x = \log r$ and $\Delta x = \Delta(\log r) = r^{-1}\Delta r$. Thus:

$$n(x) = r\nu(r)$$
$$\nu(r) = e^{-x} n(x) \qquad [1.1]$$

In theoretical work on coagulation it is especially convenient to use volume rather than radius or log radius as a size measure, but as a practical scale for displaying the distribution it is highly inconvenient. A quite dif-

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