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Introduction

1.1 Aerosols

The scientific definition of an *aerosol* refers to it as a disperse system of liquid or solid small particles suspended in a gas, usually air. It applies to a very wide range of particulate clouds encountered terrestrially, including naturally occurring airborne dusts, mists, clouds, sandstorms and snowstorms, as well as the man-made smokes, fumes, dusts and mists that are found in our working and living atmospheric environments. It also applies to airborne particles of biological origin, including pollens, spores, bacteria and viruses. Although there are some examples of beneficial aerosols, including those specifically generated for medicinal and therapeutic purposes (e.g. inhalers like those widely used by people suffering from asthma) and those that are generated in closed systems for the purpose of specific engineering applications (e.g. carbon black manufacture, nanotechnology), the primary focus of this book is with aerosols that have the potential to be harmful to human health. This alone has been the subject of great – and continuing – interest. In this regard, many aerosols are regarded as pollutants and so are considered undesirable, especially if inhaled where they may be associated with adverse health effects in humans. It follows, therefore, that much occupational and environmental health research and practice is concerned with aerosols that are regarded as hazardous agents.

Figure 1.1 summarises some aerosol classifications. It also includes some examples of aerosol types encountered in occupational and environmental hygiene, reflecting wide ranges not only of chemistry and biology but also of particle size. As indicated, the range of scale is enormous, across all of which particles have the ability to remain airborne for significant durations in most environments and so be available for inhalation by humans. It will be a consistent theme throughout this book that particle size is one of the most important aerosol indices that influences the physical nature of aerosols – including their manner of transport in the air – and in turn the intensity of human exposure and dose. By a similar token, particle size is also extremely influential in all aspects of how aerosols are measured. But composition too is an important consideration since this is largely what drives toxicological effects in aerosol-exposed people. It may vary greatly throughout the particle size range, from location to location, and from one set of environmental conditions to another. This is especially the case for ambient atmospheric aerosols, which are subject to more widely varying conditions than workplaces, where aerosol composition, although often complex, is usually fairly constant. The continuing need to understand all these aspects of aerosols poses difficult and interesting challenges to the science and practice of aerosol sampling.

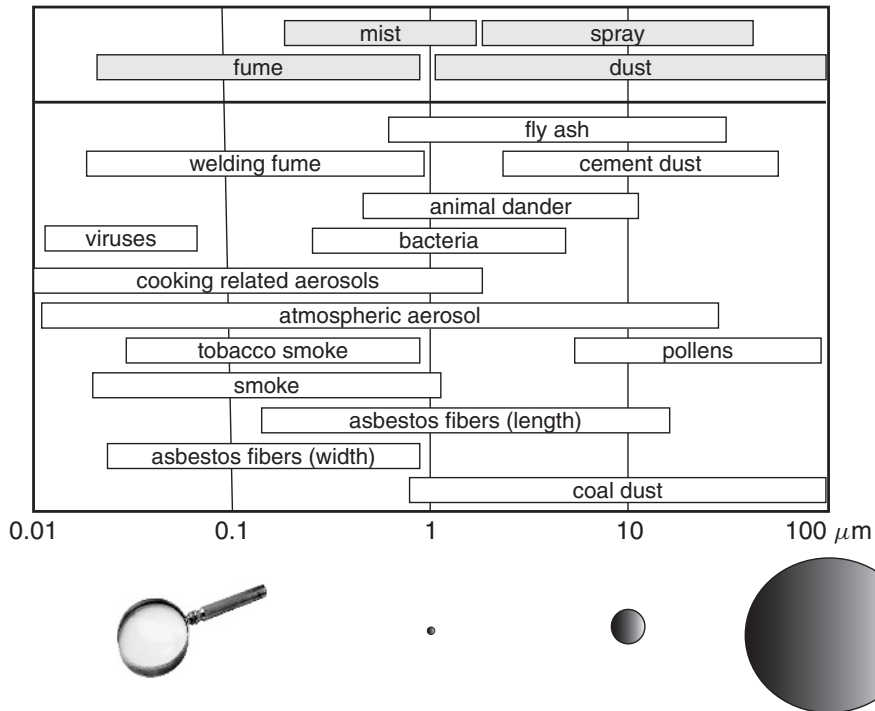


Figure 1.1 Summary classification of aerosols, some types of interest in occupational and environmental health, and an indication of the relative range of scale of particle sizes

1.2 Particle size

For a particle that is spherical, its definition is straightforward, requiring specification only of a single dimension, the particle *geometric diameter* (d). This is what would be obtained if the particle were to be observed and sized – by reference to an appropriate calibrated graticule – under a microscope. For a particle that is not spherical, however, the microscopist has a dilemma. There is no single defining dimension. Although spherical particles may be found in carefully generated laboratory aerosols, most aerosols in the real world – with the exception of mists and sprays – come into this second category. It therefore becomes necessary to find a metric of particle size that is not only consistent and accessible but is also relevant to the aspects of the behavior of the particles that are of specific interest. This leads to the concept of ‘equivalent’ particle size, for which there are several options. For example, *equivalent volume diameter* (d_V) is the diameter of a sphere that has the same volume as the particle in question, and is physically relevant to some aspects of particle motion, including diffusion. The *equivalent projected area diameter* (d_{PA}) and the *equivalent surface area diameter* (d_A), are both physically relevant to some aspects of how particles interact with electromagnetic radiation, including visible light. However, as will be seen later in Chapter 2 and beyond, none of these definitions is sufficient to fully describe the airborne behavior of the particle. From physical considerations, involving not only geometric size but also particle density and shape, particle *aerodynamic diameter* (d_{ae}) emerges as a property of great importance relevant to airborne behavior.

1.3 Elementary particle size statistics

Only rarely in practical cases do aerosols exist that consist of particles all of one size. When they do, they are referred to as ‘monodisperse’. More generally, however, aerosols consist of particles of many different sizes, with particles usually belonging to populations with statistically continuous particle size distributions. These are referred to as ‘polydisperse’. For the purposes of most of what follows in this book, a rudimentary outline of particle size statistics provides sufficient background. More detailed descriptions are available elsewhere (e.g. Fuchs, 1964; Hinds, 1999).

For particles whose size can be represented in terms of a single dimension (sometimes referred to as ‘isometric’), the fraction of the total number of particles with diameter falling within the range d to $d + dd$ may be expressed as:

$$dn = n(d) dd \quad (1.1)$$

where

$$\int_0^{\infty} n(d) d(d) = 1 \quad (1.2)$$

and where $n(d)$ is the *number frequency* distribution function. Alternatively, the *mass frequency* distribution function $m(d)$ is given by:

$$dm = m(d) dd \quad (1.3)$$

and

$$\int_0^{\infty} m(d) d(d) = 1 \quad (1.4)$$

Other forms of expression for the particle size distribution may similarly be described. They are all interrelated. Which form is actually used in practice depends on how the particle size is measured. For example, the counting and sizing of particles under a microscope yields distributions in the form of $n(d)$, while gravimetric methods, involving the weighing of collected samples on an analytical balance, yields them in the form of $m(d)$. As a general rule, when discussing particle size statistics, it is prudent to indicate the method by which the particle size analysis has been achieved.

It is often helpful in particle size statistics to plot distributions in the alternative cumulative form, for example:

$$\text{Fraction of mass with particle diameter less than } d, C_m(d) = \int_0^d m(d) d(d) \quad (1.5)$$

A typical particle size distribution is given in Figure 1.2, shown both as mass frequency and cumulative functions, $m(d)$ and $C_m(d)$, respectively. This figure contains a number of important features. Firstly, the median particle diameter, at which 50% of the mass is contained within smaller particles and 50% is contained within larger ones, can be read off directly from the cumulative distribution [Figure 1.2(b) and (c)]. Secondly, the frequency distribution [Figure 1.2(a)] exhibits a strong degree of asymmetry such that the peak value of $m(d)$ lies at a value of d which is smaller than the median. The long tail in the distribution that extends out to relatively large particles is very common – indeed is ubiquitous – in aerosols in the real world. Skewed frequency distributions like the one shown in Figure 1.2 are often well represented by the log-normal function, given for example by:

$$m(d_{ae}) = \frac{M}{d_{ae} \sqrt{2\pi} \ln \sigma_g} \exp \left[-\frac{1}{2} \left(\frac{\ln d_{ae} - \ln \text{MMAD}}{\ln \sigma_g} \right)^2 \right] \quad (1.6)$$

6 Aerosol Sampling

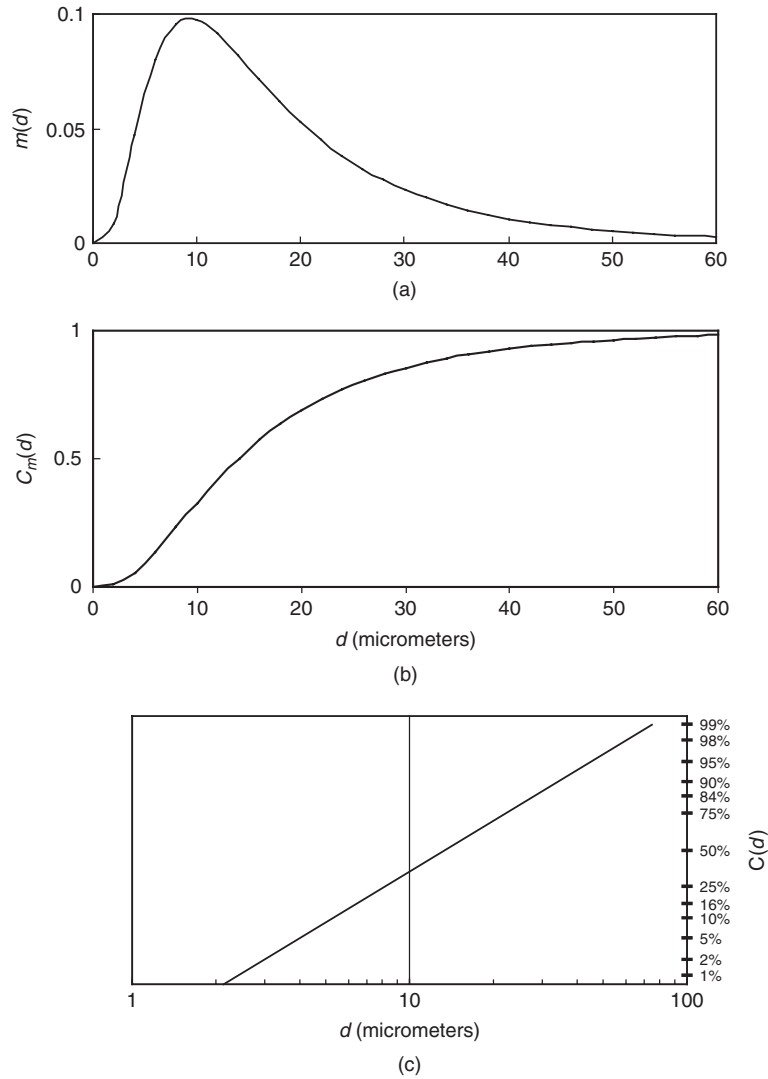


Figure 1.2 Typical aerosol mass-related particle size distribution: (a) frequency distribution; (b) cumulative distribution; and (c) cumulative distribution on log-probability axes, indicative of a log-normal particle size distribution

for the *mass* distribution in terms of particle aerodynamic diameter. Here MMAD is the commonly used acronym for the mass median aerodynamic diameter and σ_g is the geometric standard deviation reflecting the width of the distribution (or degree of polydispersity), while M is the total mass contained within the sample represented by distribution shown. Particle count and surface distributions may be also similarly represented. In Equation (1.6), σ_g is given by:

$$\sigma_g = \frac{d_{84\%}}{\text{MMAD}} = \frac{\text{MMAD}}{d_{16\%}} \quad (1.7)$$

But for calculation purposes, especially when the cumulative distribution is required, involving numerical integrations of $m(d)$, application of Equation (1.6) becomes unwieldy. The alternative expression:

$$C_m(d_{ae}) = \frac{\exp[a + b \cdot \ln(d_{ae})]}{1 + \exp[a + b \cdot \ln(d_{ae})]} \quad (1.8)$$

provides a very good approximation to the cumulative log-normal function and is much easier to use. Here:

$$b = 1.658/\ln(\text{MMAD}) \text{ and } a = -b \cdot \ln(\sigma_g) \quad (1.9)$$

For a perfectly monodisperse aerosol, $\sigma_g = 1$. More typically, for aerosols like those found in many workplace situations, σ_g ranges from 2 to 3. In such cases, when the cumulative distribution function is plotted on log-probability axes [as shown in Figure 1.2(c)], it appears as a straight line. Such behavior is easiest to interpret for scenarios where there are just single aerosol sources. But similar trends are also found for situations where there are multiple sources producing aerosols with essentially similar ranges of particle size. However, in situations where there are distinctly different types of aerosols generated into the same space, the individual particle size distributions characteristic of each source type can be clearly seen. The case illustrated in Figure 1.3 is typical of what might be found in a dusty workplace – for example, a coal mine – where, in addition to the dust sources associated with the primary mechanical mining operations, fine carbonaceous fume is also present in the form of diesel exhaust from mining vehicles. As will be seen later in this book, knowledge about particle size distribution can be extremely important in understanding the distribution of the deposition of inhaled particles to different parts of the respiratory tract. Knowledge of how chemical species are distributed throughout the overall particle size range is an important additional ingredient. Figure 1.3 therefore provides information that may be regarded as an aerosol ‘fingerprint’, containing much of the information needed for the estimation of health related dose and, in turn, risk.

The particle size distribution of an aerosol in the ambient atmosphere is highly relevant to aerosol sampling in the outdoors environment. Here, because of the very diverse range of sources together with the greater lifetime of some particles after they are released into the atmosphere, the picture is more complicated. Figure 1.4 contains a typical example, showing three clearly distinct regions. The first, the

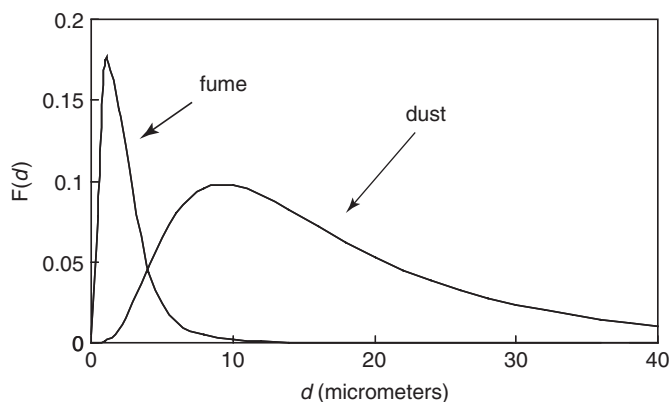


Figure 1.3 Example of a typical bi-modal particle size distribution in a workplace (e.g. coal mining) where both dust and fume are being generated

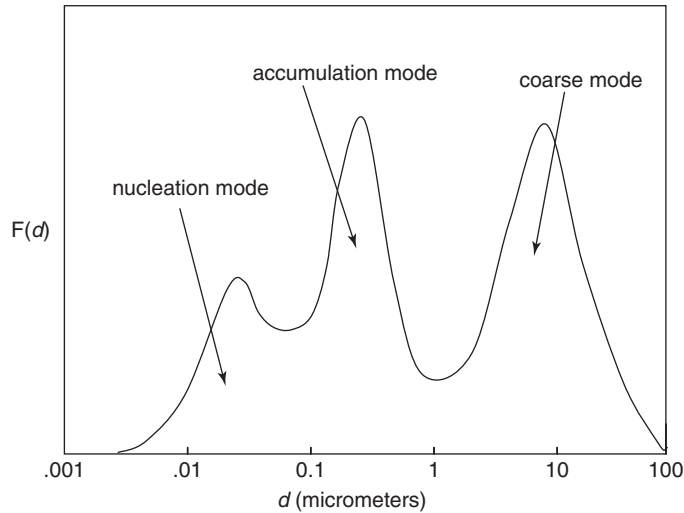


Figure 1.4 Example of a typical particle size distribution for atmospheric aerosol

coarse mode, is for the larger particles that are generally associated with mechanically generated airborne particles, including dusts from both natural and anthropological sources. The second, the *nucleation mode*, is for the finest particles that are generated as primary particles during combustion or other hot processes. Chemically, these are very different from those in the coarse fraction. In addition, following generation, they continue to evolve through both chemical and physical processes. Through some of these processes, involving condensation and coagulation, a third population of particles is produced, the *accumulation mode*. The transition between the nucleation and accumulation modes takes place for particle diameter around about 0.05 and 0.1 μm , and that between the accumulation and coarse modes around about 1.5 to 2.5 μm . As will be discussed later, these transition regions have important implications for aerosol sampling.

1.4 Aerosol measurement

Interest in aerosol sampling is stimulated by practical needs to understand, qualitatively and quantitatively, the properties of airborne particles in many occupational and ambient environmental situations. Such needs include the monitoring of emissions of particulate material to the atmosphere from industrial processes, assessment of the aerosol exposures of people for the purpose of epidemiology or risk assessment, and assessment of compliance with regulatory standards. In addition, there is the need in many situations to measure aerosols for the purpose of monitoring and controlling certain types of industrial process, including clean rooms. Interest in aerosol measurement across this range of situations expanded rapidly during the postwar period, fueled in particular by increasing public awareness of the problems associated with air pollution, especially in relation to health effects, leading in turn to the introduction of wide-ranging clean air acts, emission control limits, and air quality and occupational health standards in many countries. It has spawned a large – and still growing – body of scientific research, aimed at informing us about how to make the process of measurement as relevant as possible to the reason for making the measurement in the first place.

The property of most common interest is the aerosol mass concentration (c) defined as the mass of particulate material per unit volume of air. It is usually expressed in units of micrograms or milligrams per cubic metre ($\mu\text{g m}^{-3}$ or mg m^{-3}). Related properties of practical interest, used in some situations, include the number and surface area concentrations. Others include the distributions of particle size and chemical composition. The act of aerosol sampling involves the physical separation of particles from a given volume of the air so that they may be assessed in terms of these – and other – properties of interest.

Much of this book will be concerned with the process of aspiration whereby a known volume of the aerosol-laden air is extracted by sucking it (with the aid of pump) through one or more orifices in a solid casing which houses a *sensing region* which usually takes the form of a filter or some other substrate which may later be weighed, examined under a microscope or otherwise assayed. The aerodynamic processes by which airborne particles enter the sampler along with the air itself are – as we shall see – very complex, depending on many physical factors associated with the sampler itself as well as its surrounding environment and particle properties. The important practical question is: for given sampling conditions, how representative is the aspirated sample of the aerosol of interest in the atmospheric environment outside the sampler? In the first instance this requires definition of what is meant by ‘representative’, and this depends on the scientific rationale underlying the initial decision to carry out the sampling. Secondly, the sampled aerosol concentration and/or particle size distribution are strongly affected by the physical sampling process itself. Since this in turn is governed by physical processes dependent on particle size such that some particles will be sampled preferentially to others, the particle size distribution of the sampled aerosol will be biased and so will not properly reflect that of the aerosol at large. It follows that the measured aerosol concentration will also be biased.

The matters of *representative sampling* and the physics of the sampling process have both been the subjects of extensive discussion during recent decades. Industry and publicly funded research has been directed towards the generation of what has now become a large body of applied research involving applications of both fluid mechanics and aerosol mechanics. This in turn has enabled considerable progress towards the development of scientifically based practical methodologies.

1.5 Sampler performance characteristics

Scientific discussion of sampler performance at all levels can begin by referring to the diagram in Figure 1.5. It represents the air flow near a single-orifice sampler of arbitrary shape at arbitrary orientation with respect to a moving airstream. The sampler itself consists of an entry in a bluff body and an inner transition section leading to the sensing region, shown here as a filter on which the sampled particles are collected. The flow pattern indicated represents the motion of the air near the sampler. In particular, the limiting *streamline* – actually a *streamsurface* – is the invisible surface which separates the sampled air from that which is not sampled. To aid the discussion, the *incident plane* is defined as an arbitrary plane sufficiently far upstream of the sampler for the flow pattern there to be undisturbed by the presence or action of the sampler. The *sampling plane* is defined as the plane containing the sampler entry, and the *filter plane* defines the location of the sensing region inside the body of the sampler.

Under given particle size and air flow conditions, N_0 is the number of particles passing through that part of the incident plane contained within the limiting streamsurface. N_s is the number passing through the sampling plane, having arrived there directly whilst airborne at all times. N_r is the number passing through the sampling plane which have undergone impaction with and subsequently rebounded (or been blown off) from the external surfaces. It is important to note that both N_s and N_r may contain particles that were not included in the N_0 originally contained within the sampled air volume. Of the particles

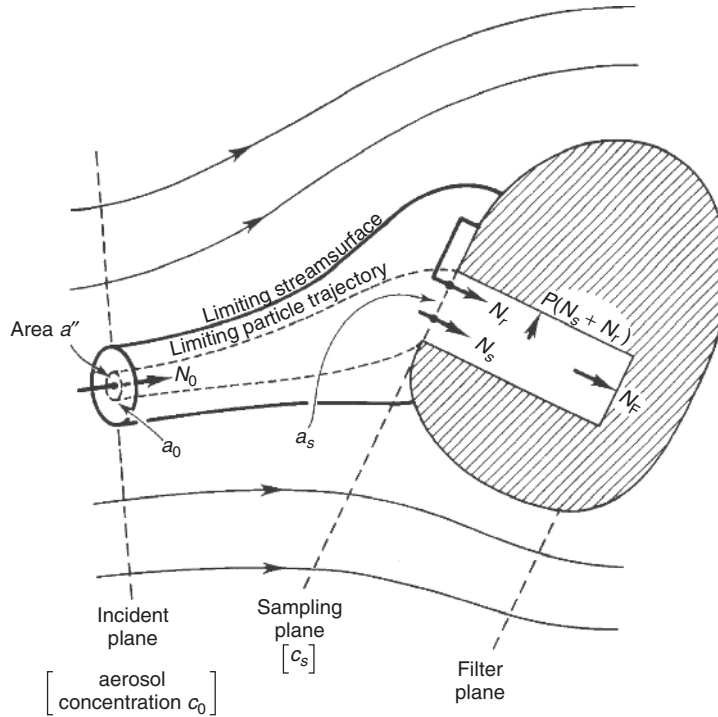


Figure 1.5 Representation of air and particle movement near a single-entry aerosol sampler of arbitrary shape and orientation with respect to the external wind, identifying the important features needed on which to base a discussion about aerosol sampler performance. Reproduced with permission from Vincent, *Aerosol Sampling: Science and Practice*. Copyright (1989) John Wiley & Sons, Ltd

crossing the sampling plane, some may be deposited on the walls of the transition section so that the number reaching the filter (N_F) is:

$$N_F = P(N_s + N_r) \quad (1.10)$$

where P is the fractional *penetration* of particles through the transition section. In what follows as this book proceeds, it will become apparent that sampler performance needs to be described by more than one parameter. Firstly, the *aspiration efficiency* (A) is the purely aerodynamic part of sampler performance, determined only by the air and particle motions outside the sampler. This is given by:

$$A = \frac{N_s}{N_0} \quad (1.11)$$

Next, the *entry* (or *apparent aspiration*) *efficiency* is:

$$A_{app} = \frac{N_s + N_r}{N_0} \quad (1.12)$$

Finally, the *overall sampling efficiency* (or *sampling effectiveness*) is

$$A^* = \frac{N_F}{N_0} = \frac{P(N_s + N_r)}{N_0} \longrightarrow \frac{PN_s}{N_0} = PA \text{ when } N_r = 0 \quad (1.13)$$

This last metric of sampler performance is of special importance when a particular fraction of the aspirated aerosol needs to be consistently selected. For example, as will be discussed in detail later in this book, this might be a sample that sets out to simulate a subfraction of particles inhaled by humans that deposit in a particular part of the respiratory tract. In such cases, the transition region between the entry and the filter is designed with regard to geometry, as well as fluid and aerosol mechanics, so that the penetration, P , is consistent and known.

Particle concentration in an aerosol is a scalar quantity that, for practical purposes (and except for extremely low concentrations), may be described in terms of a continuous spatial distribution. Consider again Figure 1.5. The limiting streamsurface encloses an area a_0 in the incident plane. The sampling orifice has area a_s , outside which all particles either strike the external wall of the sampler or pass by in the freestream. The limiting particle trajectory surface, inside which all particles enter the plane of the sampling orifice, encloses a corresponding area a'' . Only for inertialess particles do the limiting streamsurface and limiting particle trajectory surface coincide. Then $a_0 = a''$. Otherwise $a_0 > a''$. The ideal situation is the one where there are no external or internal wall effects on sampler performance, in which case Equation (1.11) for aspiration efficiency may be rewritten as:

$$A = \frac{\int_{a_s} (cv) da}{\int_{a_0} (cv) da} \quad (1.14)$$

where c and v are local values (over local elementary areas da) for particle concentration and velocity, both distributed continuously across the sampling and incident plane respectively. Here, terms of the form (cv) are local particle fluxes, the numbers of particles flowing through unit area per unit time. Since, by continuity, the net flux of particles through a'' is the same as that through a_s , Equation (1.14) may also be expressed as

$$A = \frac{\int_{a''} (cv) da}{\int_{a_0} (cv) da} \quad (1.15)$$

It is obvious from Equation (1.15) that A is dependent on the spatial distribution of both particle concentration and velocity across the incident plane unless:

$$cv = \text{constant} \quad (1.16)$$

over a_s , or a'' , whichever is the greater. When Equation (1.16) is satisfied, Equation (1.15) reduces to:

$$A = \frac{a''}{a_0} \quad (1.17)$$

and this is the form that underlies most theoretical – and some experimental – assessments of aspiration efficiency. In terms of particle fluxes, Equation (1.17) leads to:

$$A = \frac{c_s U_s a_s}{c_0 U a_0} \quad (1.18)$$

where U is the freestream air velocity at the incident plane and U_s the mean air velocity across the sampling plane, and where – as already defined – c_s and c_0 are the mean particle concentrations at the sampling and incident planes, respectively. Continuity requires that:

$$U_s a_s = U a_0 \quad (1.19)$$

So, for aspiration efficiency, Equation (1.18) becomes:

$$A = \frac{c_s}{c_0} \quad (1.20)$$

It is important to reiterate that arrival at this familiar expression is entirely dependent on the assumption of uniform spatial distributions of particle concentration and freestream air velocity upstream of the sampler.

It follows from Equation (1.13) that the efficiency by which particles arrive at the filter is

$$A^* = \frac{c_F}{c_0} \quad (1.21)$$

where c_F is the concentration of the particles reaching the filter.

In the last two expressions, c_s and c_F are the measured quantities of interest. The first depends on the aspiration efficiency of the sampler as reflected in A . This in turn is governed by the physics of the aspiration process so that for fixed geometrical and fluid mechanical variables, it is a function of particle size. As shown in Equation (1.13), the second depends both on A and on the similarly particle size-dependent efficiency P by which particles penetrate through the internal section of the sampler between the inlet and the filter.

For a polydisperse aerosol where the non-normalized mass-based particle size distribution is given by $M(d)$, c_s is given by:

$$c_s = \int_0^{\infty} A(d)M(d) dd \quad (1.22)$$

and c_F by:

$$c_F = \int_0^{\infty} P(d)A(d)M(d) dd \quad (1.23)$$

As will become apparent, Equations (1.20)–(1.23) are central to all discussions about the performance characteristics of aerosol samplers.

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