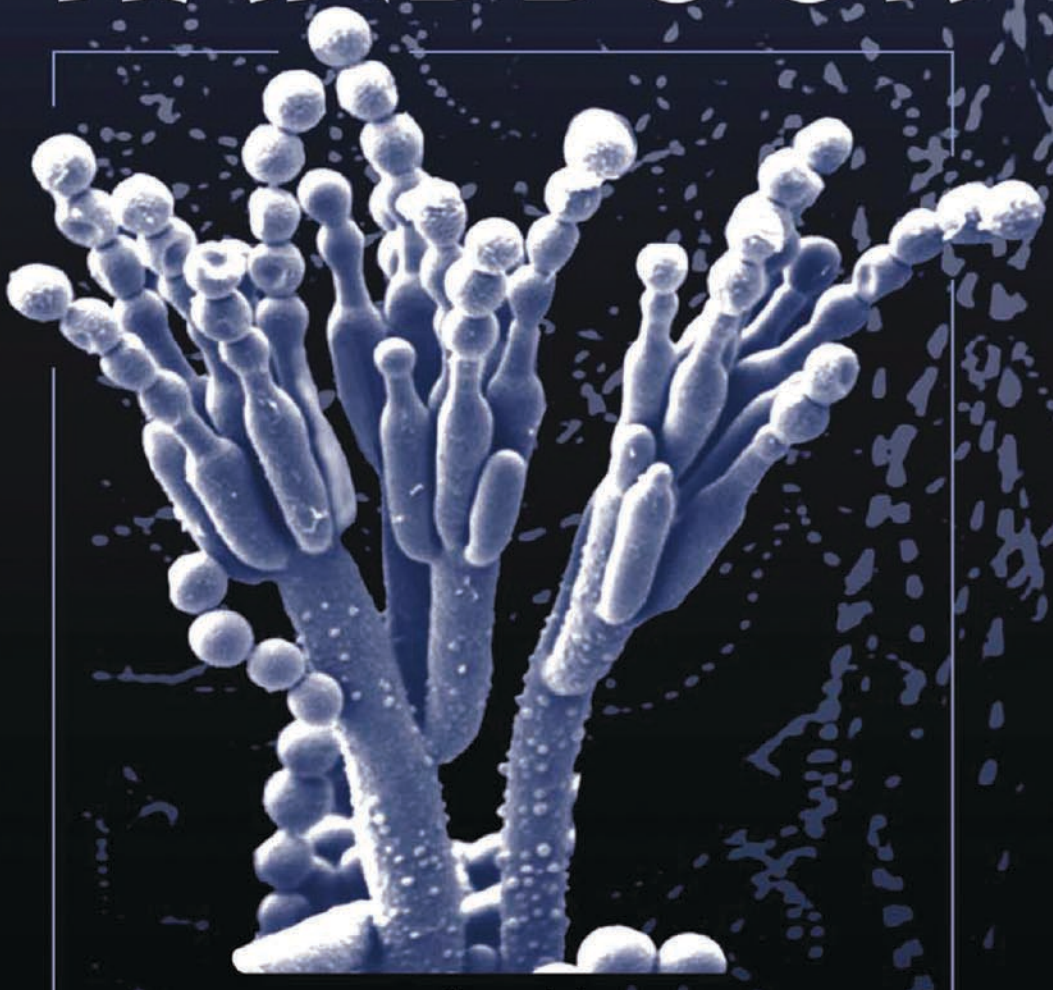
BIOAEROSOLS HANDBOOK



Edited by

Christopher S. Cox
Christopher M. Wathes

CRC Press
Taylor & Francis Group

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CRC Press
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Boca Raton London New York

CRC Press is an imprint of the
Taylor & Francis Group, an **informa** business

First published 1995 by Lewis Publishers

Published 2020 by CRC Press
Taylor & Francis Group
6000 Broken Sound Parkway NW, Suite 300
Boca Raton, FL 33487-2742

First issued in paperback 2020

ISBN 13: 978-0-367-57973-9 (pbk)
ISBN 13: 978-0-87371-615-4 (hbk)

Visit the Taylor & Francis Web site at
http://www.taylorandfrancis.com

and the CRC Press Web site at
http://www.crcpress.com

Library of Congress Card Number 94-44749

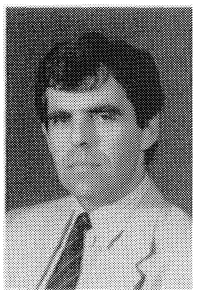
Library of Congress Cataloging-in-Publication Data

Bioaerosols handbook / edited by Christopher S. Cox, Christopher M. Wathes.
p. cm.
Includes bibliographical references and index.
ISBN 0-87371-615-9
1. Air—Microbiology—Handbooks, manuals, etc. 2. Aerosols—Handbooks, manuals, etc. 3. Air—Pollution—Handbooks, manuals, etc. I. Cox, C.S. II. Wathes, C.M.
QR101.B55 1995 94-44749
576'.190961—dc20 CIP

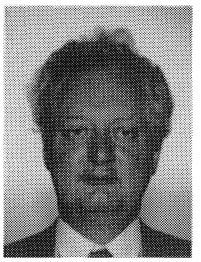
Acknowledgments

The editors are extremely grateful to the authors and publishers for their patience, perseverance and enthusiasm for this Handbook, especially Jon Lewis and Sharon Ray of Lewis Publishers. Loraine Clark, assisted by Mavis Battey and Barbara Sydnor, Silsoe Research Institute, prepared all of the chapters up to proof stage as well as carrying out many of the administrative chores. These tasks were undertaken with great efficiency and good humor, for which we are especially thankful.

About the Editors



Christopher M. Wathes is an environmental scientist, originally trained in physics. After 10 years in the Department of Animal Husbandry at the University of Bristol, England, studying aerobiology in the context of animal houses, he moved to Silsoe Research Institute in 1990 to become Head of the Animal Science and Engineering Division. He has a long standing interest in the generation, dispersion and survival of bioaerosols in livestock buildings, including their effects on animals. Currently his two major interests are emissions of atmospheric pollutants within and from animal houses; and environmental influences on animal behavior and welfare.



Christopher S. Cox was educated at Bristol University, where in 1958 he obtained a B.SC. in special Honours Biochemistry and in 1961 his Ph.D. in Biophysical Chemistry. On completing his thesis, concerned with roles of water molecules in biological systems, he joined the Ministry of Defence, UK, initially as a Junior Research Fellow at the Microbiological Research Establishment (MRE), Porton Down, UK. On its closure in 1979, he transferred to the Chemical and Biological Defence establishment. His career has included also five years working in the U.S. as a Guest Scientist of the Department of Defense. Dr. Cox's published works include two books together with about 60 research papers all concerned with different aspects of bioaerosols and microbial survival following desiccation.

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CHAPTER 1

Editors' Introduction

Many processes generate bioaerosols of diverse forms ranging from submicron allergens to much larger fungi, pollens, droplet nuclei, and dust rafts. Humans and animals are disseminators, e.g., during sneezing, while acting too as reservoirs and amplifiers. Indoor bioaerosol particles comprise respiratory pathogens, contaminated skin squames, dust mite fragments/faeces, fungal spores, hyphae and products, etc. Residential environments may present more serious risk through infection and allergy than those of lower bioaerosol concentration as occur outdoors. Reduced house ventilation rates may benefit energy conservation but concomitantly result in higher bioaerosol concentrations and risks of associated diseases. Offices, schools, hospitals and industrial workplaces, similarly are contaminated, while practices conducted therein contribute to bioaerosol burdens. Industrial workplaces provide further sources owing to contamination by, and the biological activity of, materials being handled, such as microbial and food allergens/toxins, industrial scale fermentation microbes and products, e.g., insulin. In animal houses, bioaerosols are produced from animals themselves, their foodstuffs, bedding and faeces. Within all buildings, particle diffusion and air currents ensure bioaerosols become distributed throughout and reach the most inaccessible places. But, during this process their biochemical properties (e.g., viability, infectivity, allergenicity) can be modified so that allergenic/immunological properties change and viability declines, while physical parameters such as size and shape alter.

Outdoors many processes cause bioaerosol liberation including air turbulence, spray irrigation, sewage treatment plants, breaking of waves, bursting of bubbles, and crop spraying. The purposeful release of pollen and spores provides a further example. The use and deliberate release of biological pesticides/genetically engineered microorganisms also represent potential hazards both during their application as well as manufacture in industrial fermenters. Our ability to successfully monitor and control any associated leaks seems essential while environmental impacts of these processes through colonization by released microbes, competition with indigenous species and genetic exchange through transformation also may present a hazard. Of concern too, is contamination via bioaerosols of food and pharmaceutical products especially where vaccines are concerned.

Bioaerosol hazards to man primarily arise from exposure to high concentrations or to unfamiliar forms, and comprise respiratory distress, microbial infection, allergenic reaction, respiratory sensitization, and toxicological reaction. Changing patterns of work and leisure have raised risks, while outdoors air pollution (particulate and gaseous) levels generally are increasing. Respiratory hazards, or our awareness of them, show an upsurge, one example being asthma in the UK. The costs to society of bioaerosol hazards are great, e.g., in the USA each year currently there are 250 million episodes of respiratory infection, i.e., 75 million physician visits/year, 150 million days lost from work with medical care costs of ca. $10 billion, plus loss of income of ca. $10 billion. There are too airborne plant and animal diseases, both causing substantial economic losses, e.g., respiratory diseases in pigs, exacerbated by poor air hygiene in animal houses with associated reduced feed efficiency, depressed growth rate and increased veterinary treatment, at an annual estimated cost in the UK of £20 million.

Exposure limits may be set for environments in which bioaerosols are found. In the UK, the Public Health Act (1936) and Control of Pollution Act (1990) are relevant, while in the USA, the Food and Drug Administration (FDA) sets stringent limits on levels of bioaerosols in pharmaceutical industries for example, especially when concerned with vaccine production. But, how are such limits to be monitored? Assessment requires the meaningful collection of representative samples, and their characterization. However, cursory examination of the aerosol literature often leads to confusing and conflicting perspectives on sampler choice, methodology and analytical procedures. In addition, what is the sampling efficiency and very importantly, how are samplers calibrated?

Even when these matters have been settled there are further questions concerning handling the catch to determine biological and physical characteristics, all of which can change in time and space. Microbial bioaerosols present additional special difficulties because of potential conflicts between their efficient sampling as particles and as viable entities. Furthermore, biological effects can be modified/exacerbated by simultaneous inhalation of other particles and/or pollutants, e.g., sulphur and nitrogen oxides.

Clearly, no single sampler/sampling protocol is likely to be adequate for all bioaerosols in their diverse environments. Two alternative strategies have been proposed: (1) a reference sampler (or protocol) can be specified with the advantage of consistency though there will be limitations to any one sampler; (2) performance standards may be set for a particular sampling objective against which alternative samplers (or protocols) can be tested. The latter encourages development of better, generic samplers/sampling procedures though gradual improvements may take time. Ultimately establishing performance standards for bioaerosol samplers/sampling is essential. The UK Department of Trade initiative in this area, as well as the UK Calibration Forum for Aerosol Analysis (NCFAA), are noteworthy.

The main objectives of the "Bioaerosols Handbook" are to provide up-to-date detailed descriptions, comparisons and calibration methods for bioaerosol samplers with appropriate sampling methodologies and analytical procedures. Physical and biological properties are considered from both practical and theoretical viewpoints. The Handbook represents a compilation of relevant, up-to-date knowledge and expertise of leading bioaerosol and aerosol scientists from six different countries in Europe and North America. The authors and editors are aware that there are other texts dealing with aerosols and measurement techniques. However, often these are theoretical and may fail to include essential practicalities of sampling, calibration or sample assay methodologies. This Handbook attempts to deal with the subject of bioaerosols on a broad yet in-depth basis, and provide guidance based on firm physical and biological principles plus practical experience of meaningful catching, assessing and monitoring bioaerosols encountered in many environments.

After two introductory chapters, the Handbook is divided into four parts. Chapters 3 through 6 examine the principles of bioaerosol sampling while emphasizing the essential foundations upon which good practice is built. Comprehensive descriptions of modern bioaerosol samplers, including direct reading instruments, are given in Chapters 7 to 10: many of the described calibration techniques are common to other aerosols samplers. Bioaerosols may be analyzed chemically, physically, and biologically, and current techniques are described in Chapters 11 to 15. Finally, Chapters 16 to 21 proffer the varied experiences of current practitioners of the 'art' of bioaerosol sampling in the workplace, home, specialized settings, e.g., laboratories and hospitals, and outdoors.

CHAPTER 2

Bioaerosols: Introduction, Retrospect and Prospect

J.M. Hirst[1]

THE FOUNDATIONS

'Bioaerosol' and the comparable term 'aerobiology' are compound words that describe studies relying on the interplay of disciplines primarily physical, chemical and biological. Neither the order of the syllables nor the history of the studies gives any guidance as to the precedence of the living or atmospheric component and attempts to identify which 'was the chicken and which the egg' would be fruitless; they must always be interactive and interdependent!

However, studies of how particles enter and travel in air are so characterized by diversity and diffusion that contributors and readers of the Handbook will need to recognize the same boundaries. The specialist will have no difficulty in distinguishing the scientific meaning of the word 'aerosol' from that thrust into common parlance by advertisements that lead the public to believe an 'aerosol' is the can bought at the chemist or supermarket and used to generate an aerosol of fly-spray, deodorant or the like. Scientists will support the definitions provided by the editors:

> An **aerosol** consists of material finely divided and suspended in air or other gaseous environment, with compositions as varied as matter itself.
>
> A **bioaerosol** is an aerosol comprising particles of biological origin or activity which may affect living things through infectivity, allergenicity, toxicity, pharmacological or other processes. Particle sizes may range from aerodynamic diameters of ca. 0.5 to 100 μm.

These definitions show how an aerosol can qualify to be a bioaerosol, yet they permit the great flexibility required to accommodate enormous biological diversity and to allow the subject to develop. Thus, life is not essential but the particles must have biological origin or activity. Also, sizing particles by *aerodynamic diameter* (see page 180) specifies airborne properties but allows the inclusion of great diversity in the dimensions and shape of biological propagules or fragments of organisms or their products formed mechanically

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[1]**Editors' note.**
This introduction to the Bioaerosols Handbook was written by Professor Hirst after he had been given sight of the text in draft. Through this unorthodox practice, he has gained the unique advantages of constructive comment based on hindsight and thereby has overcome the potential limitations of inadequate integration that can hamper multi-author volumes.

Inflexible definitions could also inhibit inclusion of desirable additions, for example, novel 'product allergens' comparable to those in the faeces of the house dust mite, or greater need for simultaneous study of biological and abiotic components that seem synergistic to biological activity.

Among the earliest recorded effects of aerosols and bioaerosols must have been types of occupational pneumoconioses that were mentioned in ancient Greek literature. Understanding grew slowly until the late 17th century when microscopic recognition became a reality. Thereafter study of airborne transmission accelerated, but erratically, for instance when important pathogens were shown to be transmitted by water or vectors rather than by dispersal in air. During the late 19th and 20th centuries the rate of development has become exponential, fostered by scientific progress, increased industrialization, and population growth (differentially causing either wealth or poverty), not to mention the consequences of international trade and conflicts. Later chapters show how all aspects of human activity are affected and exemplify the roles of viruses, bacteria, fungi, algae, plant microspores and many products with biological activity. The roles of many in causing the various plagues that afflict plants, animals and man are justifiably stressed in the Handbook as are the increasing concerns with the contamination of food, pharmaceutical products and effects related to environmental pollution. Currently these are the effects that most attract both interest and support. The hazard list (as exemplified in Chapter 1) is of course prodigious and very costly but judgement will not be balanced unless we also consider what may be receiving less attention than it should; has anything been neglected? In time, science will suffer if it forgets that bioaerosols are probably as beneficial as they are hazardous. Most of the benefits operate so effectively yet so surreptitiously that they are easy to overlook and although the costs of the hazards have to be met and so can usefully be estimated the same would not be possible or useful for the benefits. Nevertheless, life on earth could not survive for long without the re-mineralization of organic matter by the essential organisms of decay, many of which arrive by air. The mosses, ferns, and grasses (and many other plants and trees) could neither reproduce nor spread properly without airborne propagules. Surely not only 'green ecologists' should be concerned that there is so little attention (other than in polar regions) to the colonization of barren or environmentally altered substrates or in spreading genetic diversity (existing or engineered) through existing populations.

The real work of the Handbook begins in Chapters 3–6, each of which helps set the rules for studying bioaerosols and together they provide firm foundations, based on the wealth of knowledge and methods of aerosol science for understanding how particles behave in air and the 'ideals' of sampling. Some of these chapters may not be bed-time reading for biologists but they must persevere with the study because it is worth repeating that only thus can they build a better understanding of the proven behavior of particles in air. These chapters provide vital clues (if not always prescriptions) as to how physical principles may be applied to generate or to capture airborne particles. They not only explain the great diversity of forces that may operate but also how important it is to understand which may operate, when and on what. For example, even Brownian movement is shown to be important on sub-micron particles within the strict confines of the alveoli of the lungs. More familiar will be roles of gravitational settling, eddy diffusion, inertial impaction, filtration and other processes, here described separately but usually variously interacting in experimental conditions and during sampling both within buildings and in outdoor weather.

Gradually, but sometimes a bit erratically, principle gives way to practice with the introduction of less idealistic conditions. It is probably helpful and certainly realistic that this transfer cannot be seamless. Complex interactions between physical factors affecting

sampling and deposition often make the path difficult to follow as complications of weather, topography and vegetation are introduced as modifiers of both liberation and deposition, and how samplers should be designed to try to match these circumstances. Consideration of how to attempt to bring some ordered measurement to the complexity of composite bioaerosols illustrates how difficult this process is, yet how necessary where total aerosol content may be important. The introduction of the concept of 'dispersal unit' raises the problem of when and what is the effective unit to estimate. When is one particle sufficient, or may a clump be required for effect or infection? How much are clumps fragmented on capture? When and in what circumstances is number most important? When should it be replaced by mass, surface area or other parameters from among those now often so much easier to measure electronically? Fortunately, perhaps, biological requirements can often be more precisely defined by selective trapping methods or catch treatment allowing cultural or visual identification. These chapters foreshadow some of the myriad complications inevitable with living things, for example, the difficulty of measuring effects of dispersal and the stresses of capture on viability, which form essential knowledge to many investigations but are of no consequence to others.

It is inescapable that the Bioaerosols Handbook, having stressed the importance of aerosol science, must gradually confront the no less difficult problems posed by the biotic components. Biologists must realize that the innumerable further complexities their target organisms will present in bioaerosol studies can only, as a last resort, justify any departure from the principles of good sampling. However, I fear that later chapters will confirm that in reality such 'last resorts' are all too common. It would also be wise for biologists to recall how often history has shown that methods first used in biology and medicine were later replaced by more capable and accurate instruments borrowed or developed from work by physicists, chemists or engineers engaged in industry, military studies, aerodynamics or meteorology. This is a point in the Handbook and in research where both biologists and physicists need, yet again, to remind themselves that their disciplines must interact and be interdependent. Success depends on both groups of discipline generating a common language and understanding of mutual aims and problems. Even when this is achieved the battle may not be won without joint attention to the problems and design of experimental sampling and of manufacture. Practitioners studying bioaerosols are often remarkably dependent on anecdotal episodes and on which instruments are available. Nevertheless, experiments should be designed to permit statistical analysis whenever possible. Also some teams need to forge close cooperative links with manufacturers jointly to develop and market convenient, reliable, robust, but accurate samplers that are well matched to biological techniques yet suited to use in laboratories, factories, forests, crops, poultry houses or patients' homes.

THE TOOLS OF THE TRADE

The diversity of particles, of aims and techniques involved in sampling bioaerosols ensures that there never will be one perfect, all-purpose sampler. That allows no excuse to abandon attempts to get as close as possible. The initial chapters indicated the enormous range of particle sizes and showed how differently their behavior is affected by the many forces active and the prevailing environment.

Chapters 7 to 10 examine how that information has been or could be used for sampling and measurement. Freely exposed surface samplers are often much more complex in action than their simple structure suggests. For those who seek it there is information to guide when and for what purposes they may reliably be used. Suction samplers are as difficult

to calibrate; the first hurdle is to collect a known volume of the aerosol containing a representative sample of all airborne particles. By sampling uniform spherical monodispersed particles isokinetically in laminar air flow this perfection can be approached and there are many specialized circumstances for which it can and must remain the aim. However, in practice assorted shapes, sizes, densities, turbulent flow and gustiness create many problems, especially when suspected interactions between aerosol components seem increasingly to demand the simultaneous measurement of all. Wide ranges of size, shape and density make this difficult enough but when, in bioaerosols, they defy recognition unless grown, need to be measured over months or years, or are released intermittently in brief episodes in response to life-cycles, variable weather or treatment, then the full difficulties of measurement become evident.

Nevertheless, bioaerosol studies would have little validity without some standards. Perfection may be unattainable but at least more attempts must be made to estimate a theoretical sensitivity and to calibrate efficiency or, perhaps more realistically, estimate the deficiencies of sample collection. Once the project objectives are defined, accumulated experience must be used to help define the samplers best fitted to adopt for the particle spectra and environmental circumstances likely to be encountered. For testing to be believed, accurate and reproducible it must begin by comparing candidate samplers against near ideals using the defined conditions and standard particle spectra that are the tools of aerosol science. Unfortunately this implies access to facilities for generating standard aerosols and sampling them in specialized wind tunnels and settling chambers. Such facilities are rare and often prohibitively costly for the funding standards available to many small manufacturers and most biological projects. Desirable, indeed indispensable, though they may be, such tests should be but the first steps in a program that progressively introduces the difficulties and assesses errors of sampling real organisms in real environments.

The chapters on instrument calibration procedures (7 and 8) are essential to those who can approach perfection but no less important to those who must strive to know how far the methods they have to use will inevitably fall short. In very few instances is such information now available; at present it is difficult to do better than quote the advice in Chapter 8:

> "Whichever strategy is chosen, it is essential that due regard be given to the problem of obtaining representative samples and avoiding biases between the inlet and measurement/collection area. Viability may also be an important issue when sampling the various types of bioaerosol. The reader is encouraged to investigate both of these aspects before embarking on any quantitative sampling exercise."

The Bioaerosol Handbook will achieve much if it imprints this message on users. The need is urgent for the same degree of knowledge, skill and experience as applied to laboratory techniques to be applied to calibration of bioaerosol samplers that are to be used in 'less-than-ideal' conditions. Jolyon Mitchell provides evidence for aerobiologists to apply many of the rules but also has to make the honest if regrettable admission and exclusion that:

> "Measurements using the various types of directional and omni-directional samplers in the open environment are a separate subject in themselves."

Two succeeding chapters (9 and 10) list and comment on the inertial and non-inertial samplers used in studying bioaerosols. Both chapters give valuable, up-to-date guidance

about accepted practice. Although well referenced, the number of instruments requiring mention greatly limits the detailed comment about each. This is a pity because correct choice of sampler depends so much on the environment, the particles sought, their concentration, trap exposure and sampling time. It is no fault of the author that, regrettably but quite usually, there is much less information available about the representativeness of the sample initially collected than of the effectiveness with which particles are retained after collection. Consequently real performance is often in doubt, a reminder of the urgency of studying Jolyon Mitchell's 'separate subject.'

Experience and available information indicates that efficiency is by no means constant, small differences can sometimes have large effects. It usually is essential to consider the environment and purpose of an investigation before deciding which trap to use and where to locate it. For example, if studying a wheat crop close to harvest, the best compromise for a prolonged, general survey of its exposure to airborne fungal spores might be made with some suction sampler but very different spectra of spore size would be found depending on height within or above the crop. However, deposition on the ear and its stalk might be measured most accurately by vertical sticky cylinders where the efficiency of capture would be related to wind speeds experienced, whereas in calm air near the soil, deposition (even of small spores) on basal leaves might best be represented by horizontal gravity slides, which would be misleading elsewhere.

There can be no doubting the correctness of the dictates of good laboratory volumetric sampling; some, such as short intakes, are consistently applicable but important ones, such as isokinetic sampling, seldom seem practical outdoors. The design of most suction samplers specifies some constant intake rates and often requires some defined orientation of the sampling inlet. These and earlier chapters give many clues about the principles conferring merits or failings of the samplers listed but too seldom provide estimates of the magnitude of their probable errors. The content of the Handbook indicates that bioaerosols are increasingly studied in enclosed environments, housing, factories and hospitals, with ever more interest in smaller particles, fortunately both factors which should incur smaller collection errors. Nevertheless, much important work with bioaerosols remains out-of-doors exposed to weather or in variable enclosures dictated by the environments and activities of the organisms under study.

It may sound heretic but having established so many of the ideals, there now seems a strong need for aerosol scientists to relinquish perfection and turn some attention to realistic imperfections. Could not some be enabled to help biologists further by defining the magnitude of the errors of samplers used or developing better ones? We need to know better the magnitude of collection errors incurred by sampling widely different particle sizes (and shapes), using a range of constant intake rates through variously shaped intakes oriented differently to air movements that vary rapidly both in speed and direction. Accurate definition may seldom be possible, but results and judgements would be much more valuable if experimenters knew what trust to place in them and could admit imperfections rather than ignore them, as is too often the case.

The value of bioaerosol sampling methods finally depends on whether they are practical, give reproducible results and effectively serve the sampling objectives. At present many investigators are forced (or worse, content) to use devices supported by few facts or advertised merely on the basis of "relative performance tests" in which a new device is compared only with some earlier but equally ill-defined instrument. Many honest investigators are aware they use inefficient devices that tell lies; at best they can hope that these are reproducible and that they have learned how to interpret their results so as to usefully assist the sciences they serve.

HANDLING THE CATCH

For particular groups of organisms (e.g., pollens and some fungi) that are difficult to grow, light microscopic recognition by experts may long offer unrivaled accuracy and specificity. However, anyone who has spent many hours (even months) scanning catches under the light microscope must fervently hope for the development of less tedious but equally effective assessments. The obstacles are formidable involving great diversity in composition, amount (size, shape, number), origin, targets, viability/infectivity, and means of detection and expressing effects. Even if there could be a single, universal catching method there could be no one means of assessing the presence and expressing the effects of bioaerosol components. There has certainly been progress with many techniques, efforts that must continue. However, the diversity of particles in bioaerosols, the minuteness of genetic differences that may affect specific functional capability, their constant susceptibility to change during and after dispersal and the selectivity required by project objectives often make successful automation capable of answering many biological problems seem still distant.

The five chapters (11 to 15) dealing with assay techniques contain much evidence of progress, much hope for future development and justifiable cautions that are expressed gently because authors quote correctly but seem loath to be prophets. There is some repeat reporting, inevitable in a multi-author volume, but here with some merit by offering the reader comment from different viewpoints. Some of the advances quoted, for example, in image analysis, programmed scanning, fluorescence marking and immunology can be adopted with little difficulty and great benefit. The practicality of others will no doubt depend on the selectivity required by project objectives and by technological improvements in methods for studying the various biological components or the activity of their products. When estimation of viability or infectivity is important, many methods are suggested but there are many limitations and an obvious need for less lethal methods of capture and retention.

In many respects the viruses and bacteria must be the most difficult groups to study. They are minute, relatively featureless and need high magnification or special methods for recognition. Because they replicate only in living (or host) cells, viruses are particularly difficult to assess. Liberation and dispersal of both bacteria and fungi is complicated by important rain-actuated processes that cannot be ignored by epidemiologists but which operate independently and contribute variably to bioaerosols. Toxicity is another important result of many fungal infections of both plants and animals. The basis of allergenicity has been studied most intensively in pollens, questions posed by current research are intriguing and suggest a need for comparable investigation among fungi and bacteria. For example, if there is some real association between anemophily and allergenicity, then is this coincidental or does it indicate some functional relationship and does the same apply to other airborne particles? Most pollens are of sizes expected to be deposited in the upper parts of the respiratory system but there is now much evidence that they may cause asthma rather than rhinitis which suggests deposition further into the respiratory system than their size would suggest. How does this happen? Is it due, as often suggested, to pollen fragments? But pollen grains seem strong, resistant structures difficult to fragment (except perhaps if eaten). Could pollen allergens exist in particles formed externally on pollen grains from which they may be detached to follow some independent route to respiratory deposition? Could the symptoms be explained by transport of the allergens within body tissues? Has it been proven not to be due to the inhalation of smaller unrelated propagules that may bear similar allergens?

Throughout this volume contributors and readers are confronted with a diversity bewildering in quantity and defying generalization. Certainly the authors' duty toward completeness leaves little room for expressing preferences, even if their experience and supporting evidence could allow this. How then are new experimenters to be helped select which methods best fit their objectives? These may, for example, range from interest in the whole aerosol entering a respiratory system to attempting to assess the gene-to-gene challenge to a newly-bred crop cultivar of some newly pathogenic variant. I admit to not knowing a ready answer which seems largely a task for the future. But I am confident that those who study the Handbook carefully will be warned off many blind alleys and find signs to many worthwhile trails.

CURRENT PRACTICE

Those who have read thus far will probably have realized from my comments and examples that although I have very nearly 50 years experience in aerobiology, my active bioaerosols research was concentrated in the first 25 years, centered on the larger-spored plant pathogenic fungi, on distant dispersal and latterly on samplers.[1] This further explanation is pertinent to my comments on the final group of chapters (16 to 21)

At first it may seem disproportionate that only Chapter 16 deals with 'the great outdoors.' However, this probably truly reflects the fact that contemporary interest and support are predominantly devoted to studying problems of enclosed spaces, special risks or processes. This results from the recognition of special risks, some technological advances, and greatly stimulated public concerns with health and environment (often powerfully bolstered by legislative imperatives). Defining how to study and meet these needs together with intense media interest has demanded attention. Although medical studies began much of bioaerosol research its intensity has recently developed greatly in homes and hospitals and added extra concerns such as new allergens, pollutants, toxins and a greater interest in opportunistic attack of patients experiencing immunological suppression. Increased processing of foods, detergents and materials has created extra needs for worker protection in laboratories as did farmers' lung disease and other hazards in animal housing.

While not arguing with this allocation of chapters, my experience and personal interest gives me reason to stress the heavy duty and responsibility placed upon the authors of Chapter 16. Bioaerosols varying enormously in kind and quantity occupy much of the global troposphere. Content is sparse in polar regions and may seldom reach the stratosphere but elsewhere is much affected by faunal and floral sources, responding to climate and hour by hour to weather in particle production, liberation, dispersal and deposition. Outdoor bioaerosols are the largest in quantity, variety and importance to the global environment. Furthermore, knowledge of interchanges between enclosed spaces and open air is essential to both. Popular myth has it that the contaminant spore of *Penicillium notatum* that led Sir Alexander Fleming toward the discovery of penicillin entered St. Mary's Hospital through his open window (they are usually more common indoors than outdoors). There can be less question about the pollen grains and basidiospores I know to have been the subject of fruitless treatment or research after deposition, initially unrecognized, on slides in histology laboratories. By contrast, ventilation and air conditioning from glasshouses, animal houses or offices have proved powerful routes for transfer of pathogens of plants, animals and man. The study of pollens in palynology offers excellent examples of multiple important actions, in cross-pollinating plants, in causing allergic rhinitis and being one of the few groups of airborne propagules resistant enough to be traced to their

ultimate fate and adding evidence of great value to the historical and fossil records. Studies of outdoor bioaerosols have been the main route for accumulating the specialist botanical, mycological and agricultural knowledge essential to recognizing propagules which they have later helped to identify as causes of hitherto unrecognized problems, infections, toxins or allergens and then assist research towards control or avoidance. The history of research into farmers' lung disease well illustrates that interdependence within biological and medical disciplines is just as necessary as with the physicists!

Chapters 17 to 21 all share a primary concern with variously enclosed spaces although each is justifiably separated because of specialized conditions and purposes. The group share many features. Many of the closed environments (e.g., animal houses and some workplaces) are certainly characterizedby very dense bioaerosol concentrations; by contrast contaminant detection in sterile rooms must be designed to detect very scarce particles. Much of the work is young and still defining its standards, testing methods and developing philosophies for action. Although the variety of problems, circumstances and bioaerosol components is so great that many studies need special methods or procedures, most investigators select samplers from among those already developed and on the market, perhaps too often this is done on the basis of reputation and convenience rather than detailed inquiry or test of suitability. For example, after 40 years of use, it may be excusable that few recall that the volumetric spore traps of the Hirst, Burkard, Lanzoni, Tilak sequence retain a standard orifice designed to catch the larger-spored plant pathogenic fungi (10–50μm) and, to avoid cluttering the microscopic image with dust, **intended not** to retain many particles <5μm dia. Studies indoors often involve small particles and slow air movements, both factors that decrease errors in collection efficiency. However, significant proportions of many aerosols sampled comprise larger fragments, clumps, or transport on relatively large rafts. On the basis of information in Chapter 8, air movements resulting from heating, ventilation and activities do seem strong and variable enough in velocity and direction to cause considerable sampling error with some particle sizes, intake volumes and orifice sizes and orientations. Some contributors do cautiously assume errors but there seem to be insufficient facts on most of the samplers used to justify the general, ready assumption that such errors are negligible. The 'comforting' idea that wearing some small sampler ensures accurate measurement of personal exposure may be most in need of confirmation and instrument calibration. Large errors may also be incurred through 'overloading' samplers or, to avoid this, by too brief exposures. Much of the sub-sampling of the catch and the subsequent manipulations to identify and assess special components must inevitably introduce considerable errors. However, estimating these rarely seems to get the attention that it deserves, it is by no means easy but accurate identification and precise and replicated assay (wherever possible) are of prime importance, particularly when used to formulate or satisfy legal regulations.

The safety of air in containment systems is subject to many different configurations. The standards required are defined by the purpose—some must maintain the highest standards, others may be required only for operator protection. Fortunately, as Chapter 17 shows, this variety has been well systematized and the categories defined and linked to permissible functions. Often the design of the containment ensures fail-safe protection, but sampling is a necessary routine check of correct operation. A lot can be achieved by standard challenge procedures. Much more difficult is to produce reliable continuous testing to indicate isolated episodes of contamination, added to which such collections would be especially subject to loss of viability of the catch during the long exposures necessary or large intakes required to give a high sensitivity of detection. The subsequent chapters exhibit great diversity in intensity, source and type of challenge, very different abilities to support costly precautions or controls. Again the diversity is great, ranging from the

operating theater through food factory and home to the henhouse. Each chapter competently describes the current situation for its special circumstances, the nature of the threats and refers to their social or economic consequences. There seems to be much need for better information on the effectiveness of the samplers that have to be used to study various threats, and particularly is this necessary in relation to the perturbations of the environment by ventilation, feeding, movement and the daily rhythms of activity. Naturally attention is first given where the challenges seem greatest or perhaps least understood. Thus sick building syndromes perhaps now merit more research than the routines of hospital hygiene. There have been notable successes such as the discovery of ventilation systems as reservoirs for Legionella infection, for house dust mites (fragments and feces) being increasingly generated in centrally heated homes, foodstuffs and bedding as well as communicable diseases having been identified as hazardous to housed livestock.

OVERVIEW

The Handbook is too late to be laid as a foundation stone for bioaerosol research. Most of the pioneers are long dead; the work is too active and promising to merit a tombstone. Yet, without doubt the *Bioaerosol Handbook* provides very honest and significant milestones that deserve both congratulation and careful study. The prodigious quantity of information fulfills the definition of a 'handbook' to be both guide and manual. Were it to claim to be the 'last word,' it would be both less honest and less challenging. Careful inquiry within the well-referenced chapters and good index will allow scholars to identify where the science needs strengthening and how best to do this. Perhaps this introductory chapter may conclude with a suggestion or two?

Such is the diversity and complexity of the tasks, propagules and methods involved that nobody believes that there will ever be one perfect, all-purpose sampler or route in bioaerosol research. The physics of aerosol science has taught us much but has more to offer biologists, particularly by studying conditions that are less-than-ideal. We need devices that get as close to truth as possible but we also need to know the magnitude of errors incurred with particles of widely different size and shape. Isokinetic and into-wind sampling seem impossible counsels of perfection in gusty conditions outdoors or in the often chaotically disturbed flows within buildings. More effort is needed to establish the best attainable with omni-directional sampling at constant intake rates and defining the errors that have to be accepted with different particles and conditions. After that, solving problems depends increasingly on biologists, although they will still need help with designing gentler samplers capable of sterile handling and collecting the widest possible range of particle sizes as bulk samples (without overload). New and existing biological laboratory techniques must be used to identify and assay catches but it would be helpful if trap catches could be subsampled to provide for simultaneous sterile culture, microscopy or other tests.

Bioaerosol sampling is confronted with bewildering complexity of components, tasks, methods, extent and urgency. There will be strong temptations to attempt to calibrate speedy new techniques alongside laborious biology. However, this seems bound to be restricted to repeated tests in familiar circumstances. How can there be effective generalization between annual samples of pollen in Tauber traps in forests or the estimation of virus particles by electron microscopy of minute samples from brief catches in enclosures? Usually enlightenment must patiently await the publication and confirmation of investigations. The need for the most accurate possible assessment of the results of each observation is stressed if the wait is to be worthwhile. Some authors have estimated a detection threshold for their collection methods and bravely attempt to relate this to effect or impact.

The task is very difficult because detection must depend on the error introduced by every manipulation of each sample and then by how much repetition and statistical design can underwrite the conclusions. Cost, facilities, survival of the catch or the need for sampling unique episodes will often limit how much of what is desirable is practical. Nevertheless, there is need for more effort to convert 'single experiences' into replicated experiments whenever possible. A great advance would be made if every investigator strove to benefit from the advice of Christopher Wathes to begin their work by honestly posing and satisfactorily answering questions framed according to the names of Rudyard Kipling's "honest serving men," namely "... what and why and when and how and where and who."

Nobody is born to study bioaerosols; it is a practice so varied that recruits arrive from very different backgrounds by devious routes of interest or necessity. Such haphazard origins bring a source of strength. Similarly the Handbook authors were chosen for diverse special skills which their chapters reveal. Nevertheless, scanning chapter reference lists reveals a "Great Rift" separating subjects, sources language and thought processes that will form a serious barrier unless bridged. It seems that few physicists, chemists or engineers have been able or had opportunity to escape their disciplines enough to think 'biologically' about the environmental circumstances and components of bioaerosols. Equally there are few in medicine or biology who have the skills or facilities effectively to study and utilize the foundations of aerosol science. There have been notable exceptions—some individuals and particularly the mixed teams studying potential military threats (although, sadly if understandably, their ability to share their knowledge was for long much restricted). Even so, the contributions of these teams both to theory and practice have been and remain among the most important. Unfortunately, the teams assembled for the later emphasis on aerosols associated with atomic and nuclear problems had almost no need for a biological component.

Does this not suggest a great need and opportunity for some broad institution, perhaps a university embracing all the sciences involved, to develop a 'center of excellence' for research and advice in the bioaerosol sciences? Such an initiative must have the specialist facilities matched by grant finance available to support collaborative teams able to call upon physical, chemical, biochemical, electronic and engineering skills. The teams should also have available members with knowledge and experience in meteorology, medicine, pharmacology, agriculture, botany, zoology and the needs and contributions of these sciences.

If in addition to providing an effective guide and manual the *Bioaerosol Handbook* could stress the need for and help establish such a 'center of excellence' able to bridge the Great Rift with interdependent scientists talking a common language, then it would have set bioaerosol research on course into the 21st Century.

REFERENCE

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CHAPTER 3

Physical Aspects of Bioaerosol Particles

C.S. Cox

INTRODUCTION

Finely divided matter when suspended in air or other gaseous environments generally is referred to as an aerosol, as opposed to a sol when suspended in solvents, and may have a composition as varied as matter itself. In contrast, the term 'bioaerosol' is more restrictive and taken to mean "an aerosol of biological origin which exerts a biological action in animals and plants by virtue of its viability, infectivity, allergenicity, toxicity, pharmacological or other biological properties, with an aerodynamic diameter in the range 0.5 to 100 μm" (see Chapter 1). The restriction in terms of upper and lower particle size limits in general is set by the circumstances of bioaerosol generation.

In one important aspect all aerosol particles have a commonality, namely, their aerodynamic behavior, and so bioaerosol particles are subject to the same physical laws as other aerosol particles. In addition, though, they are subject to laws concerned with their special biological properties. In this chapter bioaerosols are considered in terms of general physical attributes, whereas biological features are subjects for later chapters.

BROWNIAN MOTION

Bioaerosol particles are bombarded constantly by molecules of the surrounding medium, which move at random. Any imparted motion is also random and the net result is that smaller bioaerosol particles perturbate about a point that drifts. This form of motion is termed 'Brownian motion' and its intensity increases with temperature and with decreasing particle size.

It may be expressed by the Einstein equation, which, in simplified form, is

$$\bar{X} = 5 \times 10^{-6} \sqrt{(t/r)} \qquad (3.1)$$

where $\bar{X}$ = root mean square particle displacement,
t = time, s
r = particle radius, cm.

So, for particles greater than about 1 μm diameter, diffusion due to Brownian motion is less than gravitational settling.

An informative example, provided by Dimmick,[1] is that because the mean diameter of the human alveolus is 15×10^{-3} cm, then for a particle to travel the alveolus radius of $7 \times$

10^{-3} cm in a 2-sec holding time, its displacement velocity would need to be greater than 3.5 $\times$ 10^{-3} cm/sec. Particles smaller than ca. 0.1 μm and greater than ca. 1 μm diameter consequently should be retained more efficiently than those within that size range.

Such is generally observed in practice.[2]

Another effect is that, owing to diffusion, very small particles tend to move away from regions of high concentration, but for the size range given above, gravitational settling is of more importance than Brownian diffusion.

GRAVITATIONAL FIELD

A particle in a parcel of still air falls owing to the gravitational field at a velocity dependent on its mass. As the rate of fall increases so does the drag or viscous frictional force acting on the particle. When the two forces are equal the particle attains its final or terminal velocity.

Stokes law relates terminal velocity to particle size, mass, etc., and can be derived by equating acceleration due to gravity with viscous drag force. For a spherical particle the terminal velocity, v (cm/sec), is

$$v = \frac{\rho d^2 gC}{18 \eta} \tag{3.2}$$

where ρ = particle density, g/cm^3
d = particle diameter, cm
g = gravitational acceleration, cm/s^2
η = air viscosity, g/cm s^{-1}
C = Cunningham slip correction.

For ambient conditions Equation 3.2 becomes,

$$v = 3.2 \times 10^5 \rho d^2 \tag{3.3}$$

so, a sphere of diameter 10 μm of unit density should fall at about 0.32 cm/sec. Such a prediction is accurate to within 1% for particles of this size but for smaller ones a discrepancy arises because as size decreases and approaches the mean free path of air molecules (ca. 6.5 $\times$ 10^{-6} cm at ambient conditions), particles slip between them. A simple form of the Cunningham slip correction factor is

$$C = 1 + \frac{2A\lambda}{d} \tag{3.4}$$

where λ = mean free path of air molecules,
A = constant of ca. unity.

Dimmick[1] suggests a useful approximation for ambient conditions, that is, to add 0.08 μm to the actual diameter before computing terminal velocity, or subtract this value from diameters calculated from such velocities. But for particles less than ca. 0.5 μm the terminal velocity is small and can be considered effectively zero.

Many bioaerosol particles have a density in the range 0.9 to 1.3 g/cm^3, and a value of 1.1 g/cm^3 is quite commonly used for computational purposes when the value has to be estimated.

A consequence of gravitational settling is that on storage in vessels particularly, and to some extent during downwind travel, bioaerosols tend to deposit on lateral surfaces. Their concentration therefore decreases with time and such losses are referred to as 'physical losses' or 'physical decay,' as opposed to 'biological decay,' which reflects loss of biological activity per se, e.g., viability, infectivity, allergenicity.

Because bioaerosol concentration can decline through both physical and biological decay processes, it is usually expedient to measure both effects independently. A common practice is to employ tracers to determine physical decay, preferably in the same experiment as biological decay. This is because physical losses can vary between experiments owing to other forces besides gravity, e.g., electrostatic, thermal gradients, etc., that can be difficult to maintain constant. Tracers have been discussed.[2]

A simplified approach is to model physical decay by a first-order decay process,

$$N_t = N_0 \exp(-k\,t) \tag{3.5}$$

where N_0 = number of particles at time t = 0,
N_t = number of particles at time t,
k = first-order decay rate constant.

And

$$\ln \frac{N_t}{N_o} = -kt$$

whence a plot of the logarithm of the airborne fraction as a function of time will be (approximately) a straight line of negative slope (-k). Values of k are functions of sedimentation behavior and chamber dimensions.

A convenient concept is that of aerosol half-life $t_{1/2}$ which is the time required for the bioaerosol concentration to become halved,

$$\ln(0.5) = k\,t_{1/2}$$

whence

$$t_{1/2} = \frac{0.69}{k} \tag{3.6}$$

This value of half-life is approximately given by[1]

$$k = \frac{v}{H} \tag{3.7}$$

where v = particle terminal velocity,
H = height of a chamber with vertical walls, or an effective height otherwise.

A nomograph[2] is available that relates particle density and half-life to size for aerosols undergoing stirred settling in a chamber having an effective height of 100 cm. Polydispersity of particle size may be allowed for by calculating the decay curve for each size class of the distribution normalized to its frequency, then a composite curve derived by addition of separate decay curves. Even though the method is relatively simple it gives results close to those observed experimentally and clearly demonstrates how polydisperse bioaerosols change particle size distribution with time owing to preferential loss of larger particles.

Over the years various attempts have been made to prolong aerosol lifetimes to beyond that achievable in a stirred settling chamber. Most have been based on a rising column of air to counter particle fall but failed because isothermal and laminar flow could not be maintained. However, an elegant and simple solution[3] was to rotate slowly a settling chamber about a horizontal axis. One way of explaining how it prolongs half-life is that particles achieve a spiral path much longer than the drum diameter. Under ideal conditions a half-life of 12,000 min has been achieved for micron-sized particles.[4]

An alternative, that is especially useful for larger particles, is to attach them to extremely fine spider escape threads.[2]

ELECTRICAL FORCES

Effects owing to electrical forces tend to receive insufficient attention, or are even ignored, in bioaerosol sampling.[5] Airborne particles on generation are invariably charged unless purposely 'neutralized,' and dry-dissemination usually generates much higher charges than does wet-dissemination. The consequences of highly charged particles include rapid aerosol mass depletion owing to enhanced surface deposition and aggregation, as well as sampling artifacts.

Aerosol discharge can be enhanced by incorporating radioactive moieties or by passing aerosols through 'charge neutralizers' containing radioactive sources. While the resulting aerosols usually have overall zero charge, individual particles are still charged with (approximately) the Boltzmann's distribution in which there are equal numbers of particles carrying charges of +1,+2,+3,+4, etc., and -1,-2,-3,-4, etc. In certain cases, though, this distribution has not been observed.[6]

The proximity of charged surfaces by induction of opposite charges in aerosol particles similarly can reduce aerosol mass and affect sampling. Usage of insulating materials in the construction of aerosol holding vessels is best avoided, therefore, while sampling devices should be conducting. Consequently, filters (e.g., Millipore) and their 'plastic holders' are best avoided unless first made conducting, e.g., by gold film deposition.[5]

Additional aspects of electrostatic charges have been considered,[2,7] while mathematical models of effects of electrostatic charges on the processes of sampling and of filtration have been derived.[8]

THERMAL GRADIENTS

Thermal gradients can be responsible for aerosol movement as particles travel from, e.g., a warmer to a colder region. When a particle is warmer on one side than the other, there is a resultant force that causes particle motion. Transparent particles usually move toward the thermal source because they act as a lens thereby focusing energy on the distal side. On the other hand, opaque particles usually move away from the thermal source and 'down' the thermal gradient.

Thermophoretic velocity depends on the material properties of aerosol particles as well as the ratio of particle size to the mean free path of air molecules. Except in thermal precipitators where temperature gradients of at least 100°C operate over a few mm or less, thermophoresis for particles larger than ca. 3 μm is slight thereby imposing an upper size limit of ca. 5 μm in sampling with such devices. On the other hand, thermophoresis provides a gentle force that helps to ensure collection of samples undamaged by thermal precipitation.

ELECTROMAGNETIC RADIATION

Aerosol particles interact with electromagnetic radiation primarily through reflection, refraction, absorption and scattering. Capture of photons by absorption can lead to re-emission at other wavelengths, e.g., fluorescence, and/or to increase in particle temperature. Heat then is lost by convection and conduction. Photophoresis occurs with non-uniform particle heating as it causes one side of the particle to be hotter. Transparent particles tend to move toward the radiation source as they act as lenses thereby focusing the radiation on their distal side. Opaque particles in contrast tend to move away from the source as the nearest side is hottest and more air molecules strike that part of the aerosol particle.

Scattering of electromagnetic radiation can be a complex process with maximum scattered intensity at 0° and 180° to the incident radiation. Elastic Raleigh scattering occurs when particles are much smaller than the wavelength and scattering intensity changes smoothly with scattering angle. (The term 'elastic' refers to when incident and scattered wavelengths are the same.) Elastic Mie scattering occurs when particle and wavelength are comparable in size, and scattered intensity changes critically with scattering angle. Even though it has been modeled only for a few simple shapes, Mie scattering forms the basis of many particle sizing instruments (see, e.g., Chapter 8).

Inelastic scattering also can occur, and the wavelength difference between incident and scattered radiation (as in Raman scattering) is characteristic of the scattering material (see, e.g., Chapter 12). Also important is particle refractive index, both real and imaginary components. The real component is equal to the ratio of the speed of radiation in air to that in the substance, whereas the imaginary component is related to the degree of absorption of radiation by that substance. Both components change independently with wavelength in a manner characteristic of the substance, while for hygroscopic materials the values depend on relative humidity as well. Consequently, derived particle sizes determined with techniques relying in some way on scattered intensity are usually not unique values. Hence, such devices need to be calibrated with the test materials themselves *under* the conditions of test (see also Chapters 7 and 8).

TURBULENT DIFFUSION AND INERTIAL FORCES

When air moves rapidly, instead of flowing smoothly (i.e., laminar flow) the flow is unstable or turbulent, and the transition between these two regimes is governed by the Reynolds number, a unitless quantity (see below). In practice, turbulence causes a random motion to be superimposed on the mean air flow. Particle aerodynamic behavior and transport properties therefore can be very different in the two regimes. This is important during transport through the atmosphere and during transit through pipes, bioaerosol samplers, sizing instruments, etc. A useful exercise is to calculate the airflow Reynolds

number in each application so that potential problems owing to turbulence can be anticipated to some extent.

In laminar flow when air carries particles, the two are usually thought of as moving together. When neither changes direction and particles are less than about 10 μm diameter this is approximately so. But, on a change of direction of air flow, the heavier bioaerosol particles having higher inertia may be unable to follow the lower inertia air molecules. Hence, bioaerosol particles may deposit on the walls of curved pipes. The conditions under which this is likely can be estimated as follows.

For a 1 μm diameter particle of unit density in a pipe 0.6 cm diameter and curve of radius 5 cm, at a flow of 10 L/min, the linear air velocity, V, is given by,

$$V = \frac{F}{\pi (\frac{D}{2})^2} \quad \text{(cm/sec)} \quad (3.8)$$

where F = flow rate, cm^3/s
D = pipe diameter, cm
hence, $V = 10^4/60 \times 1/\pi \times 1/0.3^2$
$V \simeq 600$ cm/sec

and

$$v = \frac{\rho d^2 A}{18\eta} \quad (3.9)$$

where v = particle velocity, cm/sec
ρ = particle density, g/cm^3
d = particle diameter, cm
A = acceleration other than gravitational, cm/sec^2
η = viscosity of air, g/cm s^{-1}
hence, $v = 1 \times [1 \times 10^{-4}]^2 A/[18 \times 18 \times 10^{-4}]$

and

$$A = \frac{V^2}{R} \quad (3.10)$$

where R = radius of pipe curvature, cm
hence, v = 0.22 cm/sec

For a curve of length L, the time spent in the curve is

$$t = \frac{L}{V}$$

and supposing the bend is a quarter of the circumference of circle radius R, then,

$$t = \frac{0.25 \times 2 \times \pi}{V} = 0.0135$$

Therefore, during the 0.013 spent in the pipe, the particle would continue in its original direction for that 0.013 and travel a distance of $0.013 \times v = 0.0029$ cm.

Hence, the particle moves 0.0029 cm nearer to the inside wall of the curved pipe, a distance insufficient to result in collision. But, for a 10 μm particle, the distance traveled becomes 0.29 cm and the probability of collision, p, is equal to this distance divided by the pipe diameter, viz.

$$p \approx 0.5$$

Consequently, only about half the number of 10 μm particles entering such a curved pipe may be expected to exit provided the air flow around the particle is not turbulent (see above).

The particle Reynolds number Re_p (unitless) is given by

$$Re_p = \frac{\rho_a v r}{\eta} \tag{3.11}$$

where ρ_a = air density, g/cm^3
v = particle velocity, cm/sec
r = particle radius, cm
η = viscosity of air, g/cm sec^{-1}

The ratio (η/ρ_a) is 0.15 and when v/r approaches the same value the air flow around the particle will be found turbulent, i.e., when the value of Re_p approaches unity. In the example above, for a 10 μm particle $Re_p = 0.1$ and the flow around the particle will have minimum turbulence, whereas for a 20 μm particle $Re_p > 1$ and it will experience turbulent flow.

Whether the air itself is turbulent can be deduced analogously, and the Reynolds number pertaining to that flow is given by

$$Re = \frac{\rho_a VD}{\eta} \tag{3.12}$$

where D = pipe diameter, cm.

When Re is 2 to 3×10^3 or greater, turbulent air flow is likely and in the above example is 2.4×10^3, so the flow in the pipe would be likely to be turbulent. The effect would be to increase the probability of larger particles negotiating the bend whereas for smaller particles this probability would be slightly less. Hence, a polydisperse aerosol may change size distribution when flowing along a curved pipe.

One may see that in practice straight pipes are to be preferred for carrying bioaerosols and that gentle curves are better than sharp ones. This same principle applies also in devices that rely on inertial forces, e.g., impactors, impingers, cyclones and stacked sieve samplers. For any test system, it is a useful exercise to carry out a mass balance so that the fate of all bioaerosol particles can be sensibly accounted for.

Inertial Impaction

Inertial impaction will be described in terms of the way it operates in impactors. These devices basically consist of a jet (or series of jets) that may be tapered or not, and are rectangular or circular in cross-section. Below the jet(s) is an impaction plate (Figure 3.1), and particle-laden air is sucked through the jet such that as it approaches that plate it changes direction. Entrained particles, owing to their momentum and inertia, cross the streamlines and move toward the plate surface. The distance moved (D) will be the particle velocity (v) in the jet multiplied by time, viz.

$$D = v \times t$$

where $t = L / V$

where L = length of curved trajectory
V = air velocity.

Hence $v = DV/L$,

but from above,

$$v = \frac{\rho d^2 V^2}{18\eta R}$$

so,

$$\frac{DV}{L} = \frac{\rho d^2 V^2}{18\eta R}$$

or,

$$D_m = \frac{d^2 V}{18\eta} \frac{L}{R}$$

with L/R a dimensionless quantity equal to 1.56 (or the ratio of 1/4 circumference divided by radius).

The quantity D_m is known as the Sinclair stopping distance and is the vertical distance (Figure 3.1) a particle travels after a directional change. It defines the probability of a given particle colliding with the impaction plate. In the example the larger particle has a stopping distance greater than the diameter of the exit tube, so the particle impacts, unlike a smaller particle for which the stopping distance is smaller than the exit diameter. There will be a particle size for which the collision probability is 0.5, and equal to the probability of passage. This particle size is the characteristic diameter for that particular jet under the operating conditions pertaining. One consequence is that collection efficiency of an impactor depends both on particle size and air flow.

Collection efficiency also is a function of the strength of adhesion between the particle and surface compared to dislodging forces (e.g., particle bounce and blow-off). Common practice is for the impaction plate to carry an adhesive. Even so, problems of trapping can remain and the virtual impactor is one way of overcoming such difficulties. In this device, instead of impacting onto a solid surface particles do so into quiescent air from which they are subsequently and more gently collected.

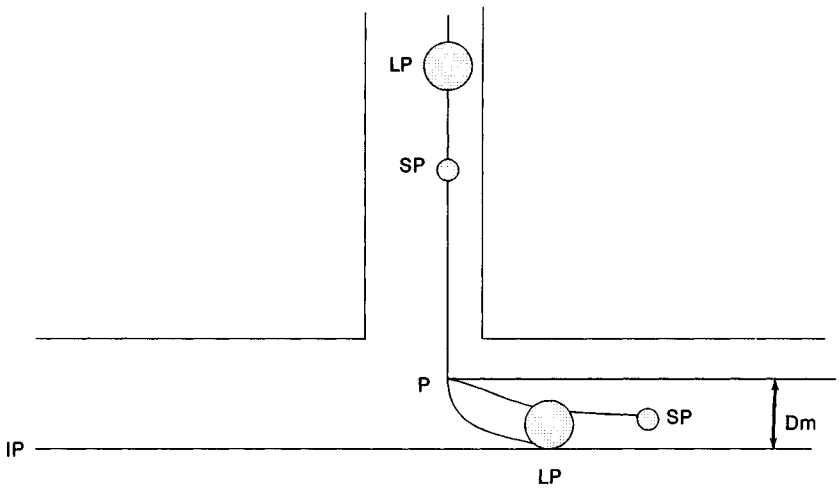

Figure 3.1. Idealized impaction in a simple jet. LP: large particle, SP: small particle, P: point of motion change, IP: impaction plate, Dm: stopping distance.

Another aspect is operating an impactor jet in the low-pressure regime. Under these conditions the pressure of the air as it exits the jet is far below atmospheric (e.g., a fine jet coupled to a high-capacity vacuum pump). At least two consequences follow: (i) the air molecules are widely separated so fine particles slip more easily between them than otherwise, and (ii) owing to the rapid expansion of the air, moisture condensation onto entrained particles can occur. Both these situations affect the collision probability of the particle with the impaction plate and can be applied to extend downward the range of particle sizes that can be sampled by impactors.

PARTICLE SHAPE

Bioaerosol particles have a wide diversity of shape ranging from spheres to spheroids to needle-like to irregular. One way of allowing for different shapes is through the concept of aerodynamic diameter, i.e., the size of a unit density sphere that behaves aerodynamically the same as the given non-spherical particle. Hence, in terms of sedimentation under gravity, two particles will fall at the same velocity even though they have different physical dimensions and density.

For sizing devices relying on particle aerodynamic behavior (e.g., impactors) this is a helpful concept especially for those applications where this is the property governing the particle behavior of interest, e.g., behavior in the respiratory tract. But, for light scattering based particle counters and sizers, etc., the concept is less helpful because the measured size depends on scattered light intensity and therefore orientation of a non-spherical particle at the time of measurement. Yet, common practice is to calibrate such instruments only with spherical particles (see Chapter 7).

There is increasing general interest in effects due to particle shape together with the availability of standard shaped particles (see also Chapter 7). For instance, these are essential for the proper evaluation of devices that try to measure both size and shape of particles.[9] One important area is how values of particle size as determined by various instruments are affected by shape, and an ensuing introduction of possible sizing errors. Another is how to formally describe particle shape and its effect on biological properties (see "Introduction") of bioaerosol particles.

One of the best ways to determine shape is by microscopy coupled to automated image analysis when formal descriptions of shape become possible.[2] But such systems can be expensive and labor intensive, hence the light scattering approaches mentioned above.

RELATIVE HUMIDITY AND HYGROSCOPY

Biological materials such as carbohydrates, proteins, nucleic acids and phospho-lipid membranes are hygroscopic, i.e., they attract water molecules. Likewise their assemblages (e.g., bacteria, viruses) are hygroscopic as are materials with which they are naturally associated (e.g., mucus). One result is that the amounts of water in these entities when airborne depend upon relative humidity (RH) and temperature. Their water sorption isotherms are S-shaped with multilayers of water molecules above about 60% RH and monolayers down to about 20% RH. They demonstrate hysteresis, i.e., there is a displacement of absorption and of desorption isotherms, and this reflects a difference in the energetics of the two processes.

Some materials undergo phase changes following dehydration, e.g., DNA, proteins, phospho-lipids, and on rehydration do not necessarily regain their original structures, e.g., protein denaturation and membrane re-organization. One reflection of this non-reversibility is viability loss (see Chapter 6).

Another consequence is that sizes of particles of such materials depend on RH, as can their shape. Of particular significance is the gross enlargement of fine hygroscopic particles on entering the respiratory tract where they are exposed to warm temperature and a water-saturated atmosphere. Whence, there is a corresponding changed deposition velocity and landing site compared to non-hygroscopic counterparts.

A further effect is that bioaerosol particle refractive index and density can be RH dependent. Then, devices utilizing such properties correspondingly have an RH- and temperature-dependent performance.

Relative Humidity Measurement

The wet and dry bulb method, or psychrometry, is probably the most common for determining relative humidity. The principle is that water, when exposed to an airstream, will cool until the vapor pressure at its surface equals that of the surrounding air. Comparing its equilibrium temperature with that of the ambient airstream permits derivation of relative humidity. In practice, provided that care is taken, the method can be both accurate and convenient. Alternatively, the dew point method provides an absolute method, but care is again required if meaningful values are to be obtained.

CONCLUSIONS

Bioaerosol behavior depends on both physical and biological attributes. Physical parameters mainly affect the where, how and in what quantities particles reach a particular location or landing site. They include diffusional, gravitational, thermal and electrostatic field effects as well as those due to relative humidity and temperature. Inertial forces and fluid dynamics are more concerned with the landing process whereas interactions with electromagnetic radiation are of interest mainly in terms of particle sizing, observation and analysis. Most if not all are functions of temperature and relative humidity.

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CHAPTER 4

Physical Aspects of Bioaerosol Sampling and Deposition

K.W. Nicholson

INTRODUCTION

The physical aspects of bioaerosol deposition and sampling are dominated by effects that relate to particle size. These incorporate the effects of inertia and sedimentation, as well as Brownian motion and sometimes electrical and phoretic effects (see also Chapter 3). Ideally, particulate materials should be sampled quantitatively, irrespective of their size, although this is difficult to perform in practice. In some cases, a knowledge of particle size is required, so that downwind air concentrations and deposition can be predicted, and this is, likewise, difficult to achieve. Meanwhile, there remain uncertainties relating to atmosphere-surface exchange rates and the interception of particles by various surface elements. The sections in this chapter aim to elucidate some of the uncertainties relating to particle sampling and transport and provide a description of the physical mechanisms involved. The reader is referred also to previous reviews in this subject area on air sampling,[1,2] particle deposition[3,4,5] and particle resuspension[6,7,8] and related material in Chapter 8.

THE PHYSICS OF PARTICLE SAMPLING

The effects of inertia usually dominate the efficiency of particle samplers, either through inlet losses or by undercollection or overcollection of particles at the inlet (see also Chapter 3). Inertial effects become increasingly important with increasing particle size and exist in any system in which there is a disturbance of the air flow. Consequently, quantitative particle sampling in the outdoor environment, in which there is a constantly changing wind flow, with regard to both wind speed and wind direction, is an especially difficult problem. Ideally, wind flow should not be disturbed and the velocity of any sampled air entering an orifice or nozzle should be equal to the local wind velocity. Such sampling is termed isokinetic (as opposed to anisokinetic) and this condition is illustrated in Figure 4.1. In areas of well-defined flow regimes (e.g., duct or stack sampling), isokinetic sampling can be achieved and the use of sharp-edged nozzles is preferred, because the body of sampler does not disturb the wind flow. In the outdoor environment, despite attempts by various workers to align a particle sampler into the wind automatically and to match the sampling velocity to the local wind speed,[9] results have been disappointing[10] and anisokinetic correction factors usually have to be applied. Such correction factors are described in many standard texts (e.g., Reist[11]) and are summarized here.

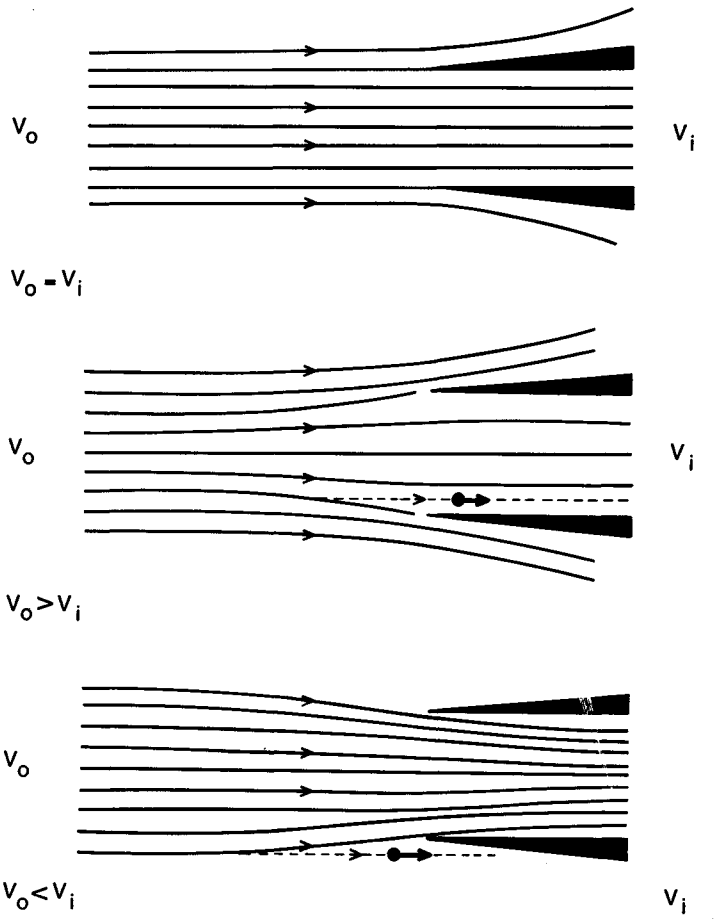

Figure 4.1. Conditions for isokinetic sampling.

A review by Stevens[12] brings some of the information together on air sampling, although much of this relates to sampling using sharp-edged nozzles. Usually, sampling efficiency is related to outside and inlet air velocities (V_o and V_i), the Stokes Number (St) and the radius of the sampling nozzle (R). The Stokes Number is defined as:

$$St = \frac{\tau V_o}{R} \tag{4.1}$$

τ, the relaxation time, can be calculated from

$$\tau = \frac{d^2 \rho_p C_c}{18\mu} \tag{4.2}$$

In Equation 4.2, d is the particle diameter, ρ_p is the particle density, μ is the viscosity of air ($\approx 1.8\times10^{-5}$ kg $m^{-1}s^{-1}$at stp) and C_c is the Cunningham correction factor. For d larger than 2λ, where λ is the mean free path of the gas molecules (≈ 0.07 μm at stp), Reist[11] approximated C_c as

$$C_c = \left[1 + \frac{2\lambda}{d}(1.257)\right] \tag{4.3}$$

Consequently, it can be seen from Equation 4.3 that in most practical cases of air sampling where inertial effects are important, C_c can be considered to be near unity.

Finally, errors due to anisokinetic operation can be estimated, according to Reist,[11] from

$$\frac{C}{C_o} = 1 - \alpha + \frac{\alpha V_o}{V_i} \tag{4.4}$$

where C and C_o are the measured and real concentrations

and

$$\alpha = \frac{2St}{1 + 2St}. \tag{4.5}$$

PRACTICAL AIR SAMPLING OF ATMOSPHERIC BIOAEROSOLS

Many practical air samplers have designs based on robustness and resistance to malicious damage, rather than on any aerodynamic considerations. In many cases, this is a necessity, since their operation may be at a remote or insecure site (see also Chapter 8). Even these designs of air sampler, however, generally are quite efficient at sampling small (less than around 10 μm diameter) particles, which are subject to limited inertial effects. A potential problem with the operation of these samplers is that their sampling characteristics, especially for the collection of large particles, usually are unknown. Furthermore, the collection efficiencies of these samplers are difficult to assess, due to the wide range and variability of meteorological conditions affecting their operation. Large particles, having the greatest inertia, have consequently the potential to act as sources of contamination, and a range of air samplers that incorporate a size-selective inlet has been designed. The upper diameter cut-off for particle entry into the sampler typically has been chosen as 10 μm, although there is no clear rationale for this decision. An assessment of performance for such size selective air samplers is relatively straightforward and can be assessed in terms of the characteristics of the inlet according to various meteorological conditions.[13] The measurement of particles smaller than around 10 μm diameter only, however, might not reflect the atmospheric concentrations of material that are available for inhalation and it is the larger particles that are most likely to be dominant in deposition processes.

Because sampling efficiency is related to the Stokes Number, an expedient for overcoming the difficulties of atmospheric sampling of large particles is to use a sampler with

large inlet dimensions. Various techniques have been developed that involve the collection of material inside a wind tunnel, which in effect, acts as a large diameter sampler. Particle collection within the wind tunnel can be well controlled and defined, while the entry of even large particles into the mouth of the tunnel is efficient. The collection techniques within the wind tunnel have included the isokinetic operation of an aspirated sampler[14,15] or the use of an impaction surface such as a cylinder[16] or a ribbon.[17] The determination of the collection efficiencies and target areas of such impactors has been studied by May and Clifford[18] and can be used to determine atmospheric concentration. Nevertheless, such samplers exist only as research instruments and have not, so far, been used in routine sampling or monitoring.

Sampling from still air (see also Chapter 8) can provide good results if the sampling velocity, V_i, is many times higher than the sentimentation velocity of the largest particles present, but not so high that inertial effects are important. Davies[19] identified two criteria:

$$\frac{V_s}{V_i} \leq 0.04 \tag{4.6}$$

where V_s is the sedimentation velocity of the sampled aerosols, and

$$St \leq 0.032. \tag{4.7}$$

Several other workers have suggested that the second criterion of Davies is too restrictive.[2,20,21] Ogden[21] reviewed the work of Yoshida et al.,[22] ter Kuile[23] and Agarwal and Liu,[24] noting the restrictive nature of Davies' criteria in relation to the results of these authors. While a number of commercial instruments fail to meet Davies' second criterion by a large margin, Figure 4.2 illustrates a range of some experimental results that demonstrate that this might not be of such major significance.

PARTICLE SIZE ANALYSIS

Typically, devices used for size fractionation studies consist of an array of impaction surfaces (see also Chapter 8). The most widely known are cascade impactors with several impaction stages, placed in series, and each stage having a lower cut-off diameter than the previous one. Each stage consists of either a single or multiple slot[25] or nozzle,[26,27] which causes particulate material to impact on a collection surface. The cut-off is determined by the velocity of air through the nozzle and the distance of the nozzle from the collection surface.

Several types of material, including glass and various metals, have been used as collection surfaces in cascade impactors. Problems of particle bounce on the initial impaction surfaces have been widely noted and this results in a distortion of the measured size range to the lower end.[28] In an attempt to overcome the effects of particle bounce, surface coatings are found to be valuable[29] and there are various greases that are commercially available. Filters placed over the impaction stages can reduce particle bounce,[30,31] although their inclusion can affect the interception characteristics of the impaction stage. May[32] has summarized a number of factors relating to impactor efficiency including gravitational effects, the occurrence of haloes and edge effects in slot impactors.

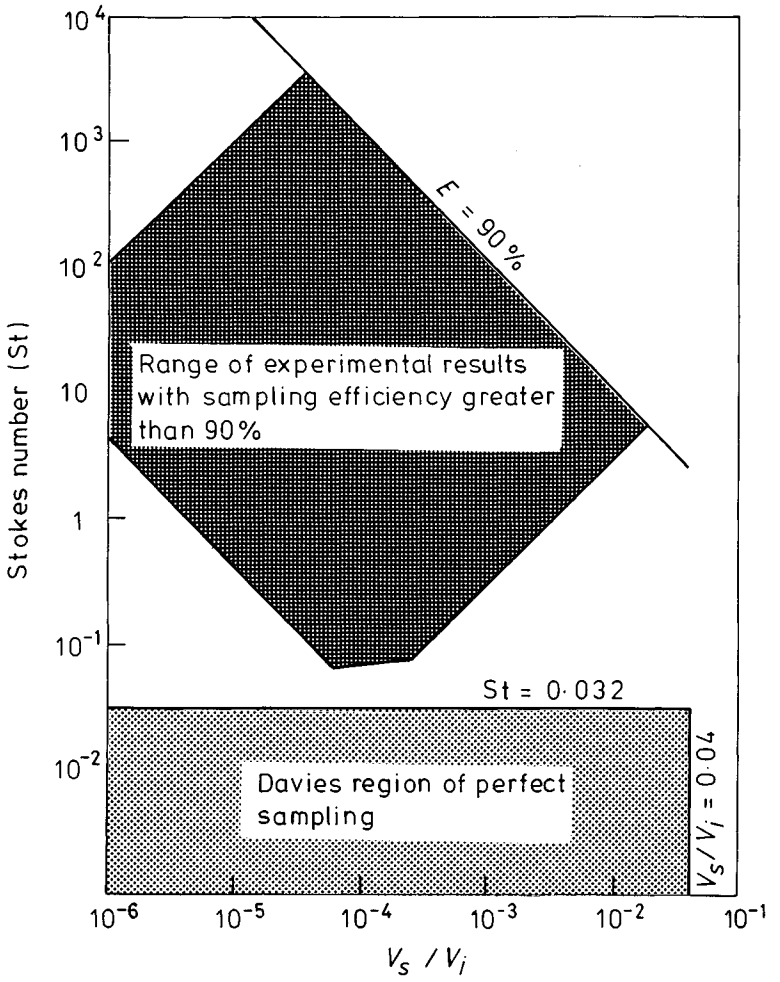

Figure 4.2. Criteria for sampling from still air (after Garland and Nicholson, 1991).

A potential problem in the use of cascade impactors is that the impaction stages can become overloaded with continual running. This may be important if lengthy sampling periods are necessary in order to measure low atmospheric concentrations or if sampling is necessary in locations of high particulate concentrations. Particle bounce from the overloaded stages may become a problem and, in order to prevent this, frequent replacement of the impaction stages is necessary. Hounam and Sherwood[33] produced a multistage virtual impactor (called a centripeter) with a high loading capacity. The principle of the centripeter is that of a traditional cascade impactor, where the impaction plate is replaced by a cup and the projected particles are captured in an enclosed void. The ability of each stage to sustain high loadings has been commented on by Yeh et al.,[34] although Biswas[35] has noted the complexity of flow in a device in which multiple virtual impactors are incorporated. Hounam and Sherwood, themselves, commented on significant wall losses within the device.

Cyclones have been used in dichotomous sampling arrangements,[36] and Liu and Rubow[37] have designed a five-stage cascade cyclone for size fractionation studies. Such cyclone devices are suitable for collecting large masses of material without overloading.

An inertial spectrometer has been designed that involves the injection of particles into a winnowing stream of air which then flows around a 90° bend.[38,39] Beyond the turn, adjacent to the outside line of the bend, is a porous plate supporting a filter medium. Sampled air is aspirated through the filter and the largest particles with the greatest inertia are sampled closest to the bend.

All of the described size fractionation devices have been found to be suitable for collecting particles up to only around 10 to 15 μm diameter.[40–42] For the size fractionation of larger particles, several devices have been proposed, although their performances in the outdoor environment have yet to be fully assessed.

Vawda et al.[43] used the principle of the wind tunnel sampler described above to collect large particles. They collected size fractionated samples, by operating a cascade impactor within a wind tunnel. Other workers[16,17] have used different impaction surfaces in various designs of wind tunnel, to get some indication of the size distribution of large particles.

Burton and Lundgren[44] described the **W**ide **R**ange **A**erosol **C**lassifier (WRAC sampler), which is designed with the intention of satisfying the criteria of Agarwal and Liu.[24] The entrance to the instrument is a 2 m long, 60 cm diameter vertical duct with a 1.5 m diameter rain shield. Size selection is achieved by four impactors operating in parallel and an additional sampler which draws air directly through a filter. The flow down the duct is controlled at 2.3 m s^{-1} and this effectively acts as a wind tunnel in which the samplers are enclosed. The large dimensions of the sampler suggest that the efficiency for large particle collection should be high, although the extreme size of the WRAC sampler means that calibration tests in a wind tunnel are impractical and are very difficult in the field.

An alternative to the sampling of particles by aspiration through an inlet, is by their collection on a rotating surface that operates in the free atmosphere. According to May et al.,[10] such a rotating device, termed the Rotorod, was first described by Perkins in 1957 (see also Chapter 9). This device consists of a square section rod which was bent in a 'U' shape and attached to a shaft of a small 12 V electric motor. When operating, the arms sweep through the air at 10 m s^{-1} and all particles larger than approximately 10 μm are impacted onto the leading surfaces of the rod, which should be coated with an adhesive. The Rotorod has been employed as a secondary standard in field experiments including other types of samplers by May et al.[10] and Vrins et al.[45] Larger rotating rod samplers have been utilized by Jaenicke and Junge[46] and Noll.[47,48] Jaenicke's device consisted of an arm with glass plates, 1 cm and 4 cm wide, rotating around a horizontal axis at 375 r.p.m. With a 30 cm radius, the glass plate passed through the air at 11.7 m s^{-1}. The axis of rotation was pointed into the wind by a vane and the plates were covered with a high viscosity silicone oil. The 50% (lower) cut-off diameters for the two plates were calculated to be 7 μm and 14 μm, respectively. Noll's sampler consisted of four sizes of impaction surface, assembled with four different radii on a rotating arm. The complete array of 16 collectors covered a range of cut-off diameters between 6 and 108 μm. Noll and Fang[49] developed a simplified device with four sizes of surface positioned on four rods of equal lengths, giving size selection over a range 7–60 μm.

Dust deposition gauges have been used to measure the deposition of large particles with significant sedimentation velocities. Usually, the gauges are employed to assess local pollution and consist of an up-facing collection disc and a drainage collecting bottle.[50] The gauges usually are not protected from precipitation and, consequently, measure material deposited in rain as well as dustfall. It is important to note, however, that such deposition gauges are suitable only for the collection of large particles and it is difficult to determine

their collection characteristics, so that atmospheric concentration and deposition to surrounding surfaces are impossible to quantify from measurements made using them. Dust deposition gauges incorporating one or more vertical slots[51,52] have been utilized to assess the horizontal flux (i.e., product of wind speed and air concentration) of material from a source. Ralph and Hall[53] have measured the collection characteristics of this type of gauge noting a 25% collection efficiency for 87 μm diameter particles and an increasing collection efficiency with increasing particle diameter. While deposition gauges, designed to measure either horizontal or vertical flux, provide a cheap and simple measurement technique, they are only efficient at collecting large particles (at least several tens of micrometers in diameter). Many environmental aerosols are too small to be effectively collected by such gauges. Although collection of depositing material by a gauge might give some indication of the deposition flux to surrounding surfaces, it is unlikely that the results would, at best, be more than semi-quantitative.

Generally, the quantitative collection of large particles requires a specialized technique. There are few commercial instruments specifically designed for this purpose and their use, anyhow, is restricted to research. The greatest problem in the operation of many routine air samplers might be due to their unknown collection efficiencies for large particles and the inclusion of a size selective inlet may be a suitable, although less than fully satisfactory solution to this problem. The application of correction factors, for anisokinetic operation in the outdoor environment, is fraught with difficulties because of the complexities of wind flow, including turbulence.

THE ATMOSPHERE-SURFACE EXCHANGE OF PARTICULATE MATERIALS IN THE OUTDOOR ENVIRONMENT

All atmospheric aerosols undergo exchange processes with outdoor surfaces. Such processes include removal or deposition from the atmosphere and resuspension or re-entrainment from underlying surfaces. Removal can occur via wet or dry deposition processes. Dry deposition is the direct interaction of a material with a surface and this process occurs continuously. Wet deposition comprises the removal of material from the atmosphere in any falling hydrometeors, such as raindrops, snowflakes or hailstones. The interception at the surface of fog and cloud droplets, which are not sufficiently large to fall readily under gravity, is a process that is not categorized easily as either wet or dry. Since these droplets are not collected efficiently by precipitation collectors, the deposition of atmospheric materials associated with them sometimes has been termed 'occult deposition.' The processes of atmosphere-surface exchange are illustrated in Figure 4.3.

The relative importance of wet and dry deposition reflects the influence of meteorological conditions, the proximity of the receptor region in relation to the source and the nature of the atmospheric material. Dry deposition is non-episodic and will occur during intervals of precipitation, as well as during dry conditions. The occurrence of certain meteorological conditions (e.g., high wind speeds) might be expected to result in higher than average dry deposition rates and the nature of the surface onto which dry deposition is occurring also is an influencing factor. Wet deposition, on the other hand, is episodic and generally increases with increasing distance from a source as materials become mixed within clouds. The wet deposition of atmospheric materials therefore can be described as resulting from either in-cloud or below-cloud scavenging. The processes of wet and dry deposition are considered in the following sections.

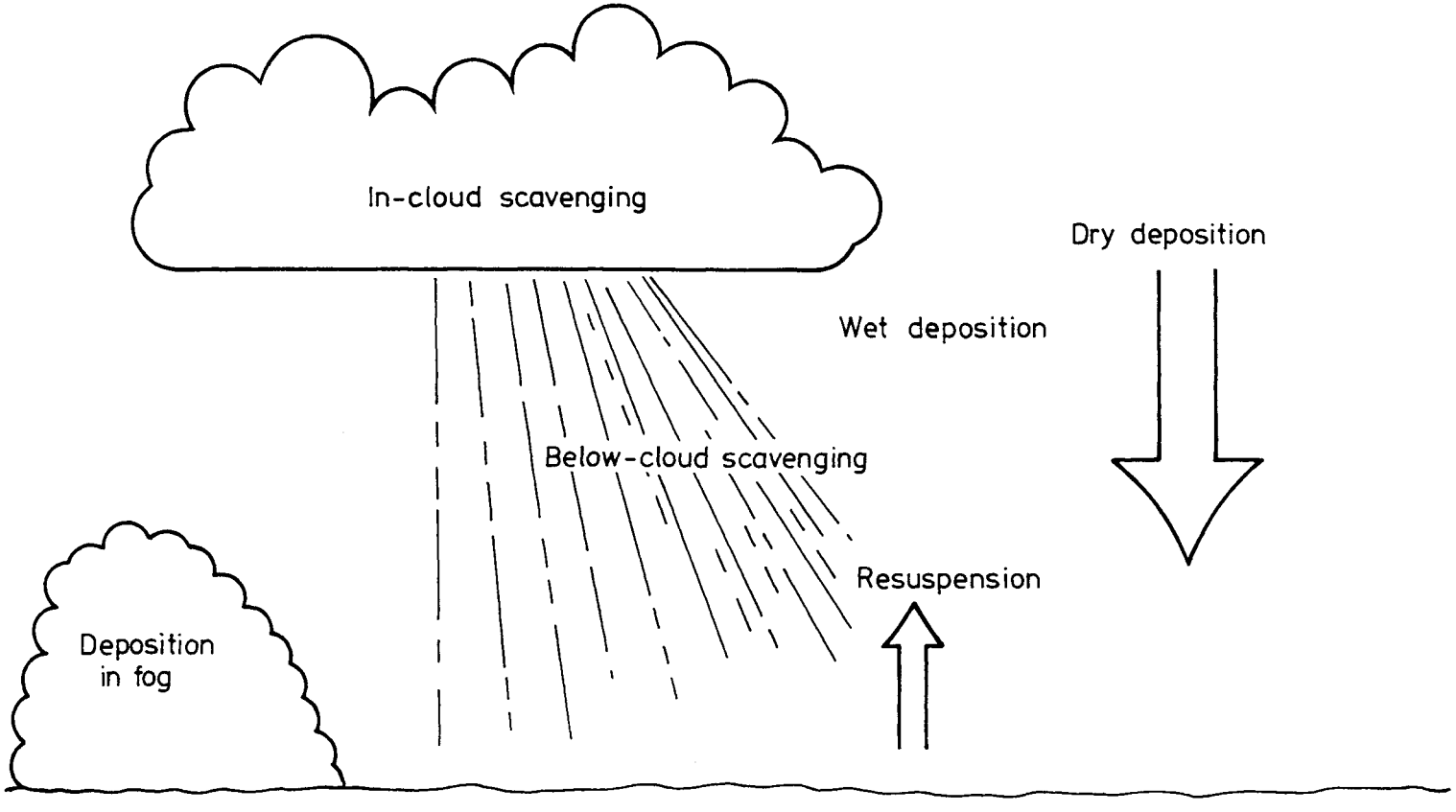

Figure 4.3. Atmosphere-surface-exchange processes.

DRY DEPOSITION AS A REMOVAL PROCESS

Dry deposition usually is expressed as a deposition velocity,[54] V_g, defined as:

$$V_g = \frac{\text{downward flux}}{\text{atmospheric concentration}} \tag{4.8}$$

The dry deposition velocity is a measure of the rate of atmospheric removal processes and is considered to be independent of atmospheric concentration for particles (although not necessarily for gases). Deposition velocity is useful for the prediction of deposition flux in the constant flux layer, which exists above any surface, the depth of which depends on the undisturbed upwind fetch and might typically extend to a few meters above the top of the surface elements, given an upwind fetch of some hundreds of meters. A concentration gradient exists in the constant flux layer and, hence, deposition velocity is a function of height. The reference height to which reported deposition velocities are quoted is somewhat arbitrary, but typically might be 1 m above the underlying surface.

To consider the atmospheric regimes that affect dry deposition, it is useful to describe the types of boundary layer that can be built up over a surface. The planetary boundary layer can extend up to 1 or 2 km, although it may only extend up to 100 m or less at night. In this layer, flow is influenced directly by large-scale surface characteristics, thermal buoyancy is significant and mixing is important. The lower part of this layer is termed the turbulent surface layer and may extend up to about 100 m during the day and, like the planetary boundary layer, subsides diurnally, to perhaps as low as several meters at night. Vertical mixing in the turbulent surface layer is controlled by the surface elements and convection. The lowest atmospheric layer, which underlies the turbulent surface layer, is described as the laminar sub-layer. This, usually at most of the order of a millimeter or so in depth, is relatively slow moving and transport to the surface is impeded by limited vertical mixing. In reality, the laminar sub-layer periodically is penetrated by turbulent eddies and it is these that may be dominant in controlling the deposition of, especially, small particles. Nevertheless, the description of the atmosphere in terms of turbulent and laminar sub-layers is a useful way of describing deposition.

In the turbulent surface layer, eddy diffusion controls transport and sedimentational effects often can be considered to be small. Like molecular diffusion, turbulent diffusion is driven by a concentration gradient, and the level of turbulence, dependent on wind speed and surface roughness, determines the efficiency of transport. It is important to note that while certain conditions might favor turbulent diffusion, transport through the laminer sub-layer can dominate particle deposition and this will be reflected in the concentration gradient in the turbulent surface layer.

The deposition flux of particulate materials in the turbulent surface layer can be equated to a diffusion coefficient K_p, where

$$\text{flux} = - K_p \, dc/dz + V_s c \tag{4.9}$$

dc/dz is the vertical concentration gradient and $V_s c$ (the product of atmospheric concentration and sedimentation velocity, see Equation 4.21) is included to allow for the effects of sedimentation. The negative sign is included on the right side of Equation (4.9) because

the deposition flux is in the opposite direction to the concentration gradient. There are a number of analogies that have been drawn between heat, momentum and particle or gas diffusion. In thermally neutral atmospheric conditions (i.e., when thermal buoyancy or damping effects are small), it is assumed generally that the eddy diffusivities of momentum (K_m), heat (K_h) and gases or particles (K_p) are identical. Here:

$$K_m = K_h = K_p = ku_* (z-d) \tag{4.10}$$

where k is the von Karman constant (≈0.41), z is height above ground level, d is the zero plane displacement (usually taken to be around 0.7 of the individual surface element height, h) and u_* is the friction velocity. u_* is a measurement of the vertical gradient of horizontal wind speed and is approximately equal to the tangential velocity of turbulent eddies within the atmosphere. The variation of wind speed, u, with height, z, above a surface can be given in thermally neutral conditions as:

$$u(z) = \frac{u_*}{k} \ln[(z-d)/z_o] \tag{4.11}$$

where z_o is the roughness length and is of order 0.1 h. In practice, u_* usually is found from the gradient of a plot of u against ln(z–d), where z_o is determined from the intercept. In such a plot, d is taken either to be equal to 0.7 h or is determined from an iterative approach.

Outside thermally neutral atmospheric conditions, the logarithmic wind profile must be corrected for the effects of convection or damping.[55] A dimensionless shear of momentum (ϕ_m) is introduced, which is defined as:

$$\phi_m = \frac{k(z-d)}{u_*} \frac{du}{dz} \tag{4.12}$$

A similar term (ϕ_h) also is introduced into the equations for heat and gas transfer:

$$\phi_h = \frac{-k(z-d)\rho u_* C_p}{H} \frac{dT}{dz} \tag{4.13}$$

where ρ = density of air, C_p = specific heat capacity of air at constant pressure, H = sensible heat flux and dT/dz is the vertical temperature gradient. The equality of K_h and K_p (but not K_m) generally is assumed, so that Equation (4.10) then can be rewritten:

$$K_h = K_p = ku_*(z-d)/\phi_h \tag{4.14}$$

ϕ_m and ϕ_h usually are determined from empirical relationships with the Richardson Number (Ri), where:

$$Ri = \frac{g\frac{d\theta}{dz}}{T\left(\frac{du}{dz}\right)^2} \tag{4.15}$$

g is the acceleration due to gravity, T is the absolute temperature and $d\theta/dz$ is the potential temperature gradient. Alternatively, ϕ_m and ϕ_h can be related to the Monin Obukhov length, L, where:

$$L = - \frac{u_*^3}{(kg\ H/\rho C_p T)} \tag{4.16}$$

(see, for example, the review by Pasquill and Smith[56]). Typically adopted empirical relationships are those of Dyer and Hicks[57] for unstable atmospheres, and of Webb[58] for stable atmospheres:

$$\text{(stable) } \phi_m = \phi_h = (1 - 5.2Ri)^{-1} = (1 + 5.2\ z/L) \tag{4.17}$$

$$\text{(unstable) } \phi_m^2 = \phi_h = (1 - 16Ri)^{-0.5} = (1 - 16\ z/L)^{-0.5} \tag{4.18}$$

Garland[55] combined Equations 4.9, 4.12 and 4.14 and equated the deposition velocity as:

$$V_g = k^2 \left\{ \frac{dc/d\ln(z-d).du/d\ln(z-d)}{\phi_h \phi_m c} \right\} \tag{4.19}$$

The above relationships between deposition flux and concentration gradient have led to a series of experiments in which deposition flux is determined from measurements of the vertical gradients of wind speed and concentration.[59–61] In practice, the deployment of such techniques for measuring particle deposition is, at least, extremely difficult.[3] Other measurements of particle deposition have been based on measurements of the covariance of instantaneous measurements of vertical wind speed (w) and atmospheric concentration (c).[62–64] Here:

$$\text{deposition flux} = \overline{w' c'} \tag{4.20}$$

where the prime denotes values above or below the mean value. The method of measurement, termed the eddy correlation method, requires rapid response instrumentation, however. It is important to add that both measurement techniques are based on turbulent

diffusion and if the particles have a significant sedimentation velocity, this will not be reflected in the measured deposition velocity.

As previously noted, particle transport through the laminar sub-layer can be dominant in controlling deposition and this is strongly dependent on particle diameter. For small particles ($\leq$ 0.1 μm diameter), Brownian diffusion is the dominant transport mechanism, and this increases with decreasing particle diameter. For large particles (> 5 μm diameter), sedimentation is an important transport mechanism, and this increases with increasing particle diameter. The sedimentation velocity (V_s) can be calculated from:

$$V_s = \tau g \tag{4.21}$$

where τ is the relaxation time as given in Equation 4.2. Equation 4.21 can be derived by equating the force on the particle due to gravity to the drag force calculated from Stokes' Law and holds true for particle Reynold's Numbers (Re_p) less than unity, where:

$$Re_p = \frac{d\ V_s\ \rho_p}{\mu}\ . \tag{4.22}$$

In practice, this means that sedimentation velocities of particles up to around 80 μm diameter can be calculated this way. As in the case of air sampling (see "The Physics of Particle Sampling"), the Cunningham correction factor should be included in the calculation of the sedimentation velocity of small particles, although this is a minor consideration in the practical applications of bioaerosol deposition. For values of Re_p greater than unity, V_s can be calculated[2] from Equation 4.21, where τ is given by:

$$\tau = \left(\frac{d^2\ \rho_p}{18\mu}\right)\ \left(\frac{24}{C_D\ Re_p}\right) \tag{4.23}$$

where

$$C_D = \frac{24}{Re_p}\ \left(1 + \frac{3}{16}\ Re_p\right) \tag{4.24}$$

Equation 4.24 is valid for $1 < Re_p < 5$, and for $Re_p > 5$ the calculation of V_s is more complicated. $Re_p < 5$, however, represents the normal range of atmospheric aerosols.

The deposition velocities of aerosols usually are considered to be greater than their sedimentation velocities. While there are considerations of particle bounce-off and blow-off from surfaces, most theoretical and semi-empirical considerations have V_g always greater than V_s (e.g., Sehmel[7]).

The declining importance of Brownian motion with increasing particle size and the increasing importance of sedimentational effects, result in a 'V' shaped curve of V_g against particle diameter, with the minimum for particles in the range 0.1 to 1.0 μm diameter (see Figure 4.4). In this intermediate range inertial effects can dominate and individual particles may impact on a surface. This may arise because the particles acquire sufficient momentum from the free-stream turbulent eddies that they are able to cross the laminar sub-layer of the underlying surface. This process is termed turbulent inertial impaction. Alter-

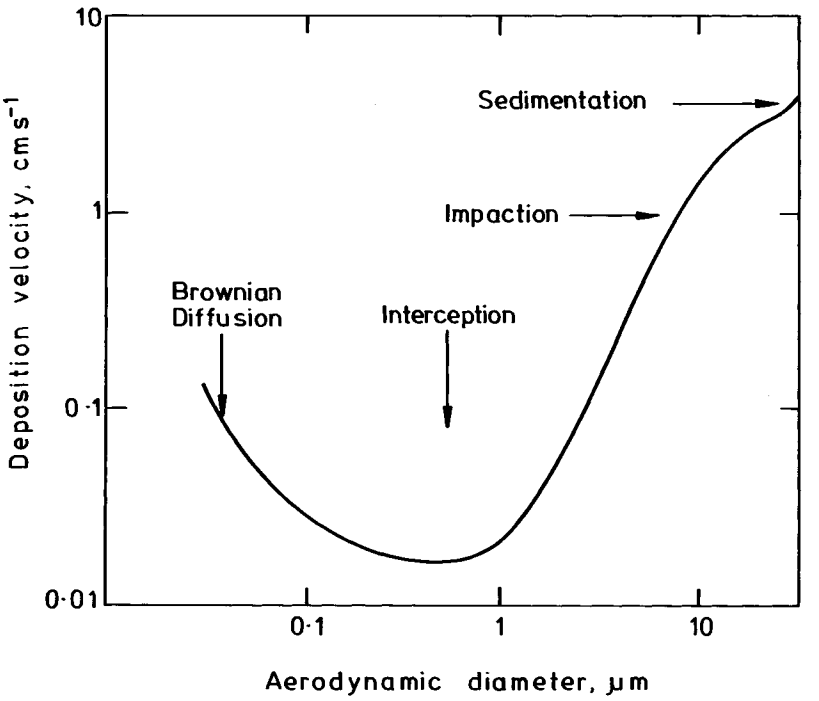

Figure 4.4. Schematic representation of deposition velocity as a function of particle size (note: values of deposition velocity are strongly dependent on environmental conditions).

natively, as is quite often the case when considering natural surfaces, part of a surface element may protrude into the free air stream and particles might impact it due to their momentum gained from movement with the wind. Particles also might be intercepted by surface elements protruding into the free air stream. Such interception occurs when a particle passes so closely to a surface element that it touches it, although it does not have sufficient inertia to cross the streamlines around the surface element. The processes of impaction and interception are illustrated in Figure 4.5.

It is for the intermediate particle diameters, not strongly influenced by sedimentation or Brownian diffusion, that the greatest uncertainties in V_g lie.[3] Both theoretical and semi-empirical predictive models have predicted values of V_g that have been near 0.01 cm s^{-1} for particles in this size range.[65–67] These values have to some extent been supported by laboratory experiments,[68–70] while field measurements of dry deposition have given anything from slightly higher values[60,63,71] to deposition velocities up to 1 cm s^{-1}.[61,72,73] The differences between the results of the laboratory and field measurements have been attributed to the different scales of atmospheric turbulence. The greater differences between the theoretical predictions and field measurements, however, probably reflect the over-simplistic approaches to modeling. Since passage through the laminar sub-layer is likely to present the greatest resistance to particle deposition in the intermediate size range, the calculation of its characteristics is crucial in determining the predicted deposition velocity. While inertial impaction onto rigid cylinders or plates can be calculated in terms of Stokes Number, the extrapolation of such calculations to outdoor conditions, where surface elements are complex in structure and may themselves not be smooth, is likely to

be speculative. The fact that surface elements might move in the wind provides another complicating factor.

A resistance analogy often has been quoted for gases[55,74] to model their dry deposition, and can also be useful for describing particle deposition. In this approach, the total resistance to transport through the turbulent surface layer (r) is considered to consist of three components: r_a, the resistance to transport through the turbulent surface layer, r_b, the resistance to transport through the laminar sub-layer and r_c, the resistance to surface uptake. r is given as the reciprocal of V_g.

$$1/V_g = r = r_a + r_b + r_c \tag{4.25}$$

For particles, the total resistance to transport (r_p) includes the effects of sedimentation, so that:

$$1/r_p = 1/(r_a + r_b + r_c) + 1/r_g \tag{4.26}$$

where

$$1/r_g = V_s \tag{4.27}$$

Generally, r_a is assumed to be equal for heat and for particles, so that in neutral atmospheric conditions,

$$r_a = u/u_*^2 \tag{4.28}$$

Outside thermally neutral conditions, an empirical correction term (ψ_c) is included. Arranging Equation 4.28 with Equation 4.13 gives:

$$r_a = \frac{1}{ku_*}\left(\ln \frac{z-d}{z_o} - \psi_c\right) \tag{4.29}$$

r_b can be related to the sub-layer Stanton Number (B)

$$r_b = \frac{1}{Bu_*} \tag{4.30}$$

where B is determined empirically and the final resistance to transport (r_c) is assumed to be equal to zero for particles. There remains the possibility of particle bounce,[75] which means that r_c is not necessarily equal to zero, although the presence of surface moisture, leaf hairs or surface stickiness would help reduce this effect.[76]

WET DEPOSITION AS A REMOVAL PROCESS

Removal of material from the atmosphere in precipitation differs from dry deposition in that precipitation is episodic and occurs over a limited fraction of time. Nevertheless,

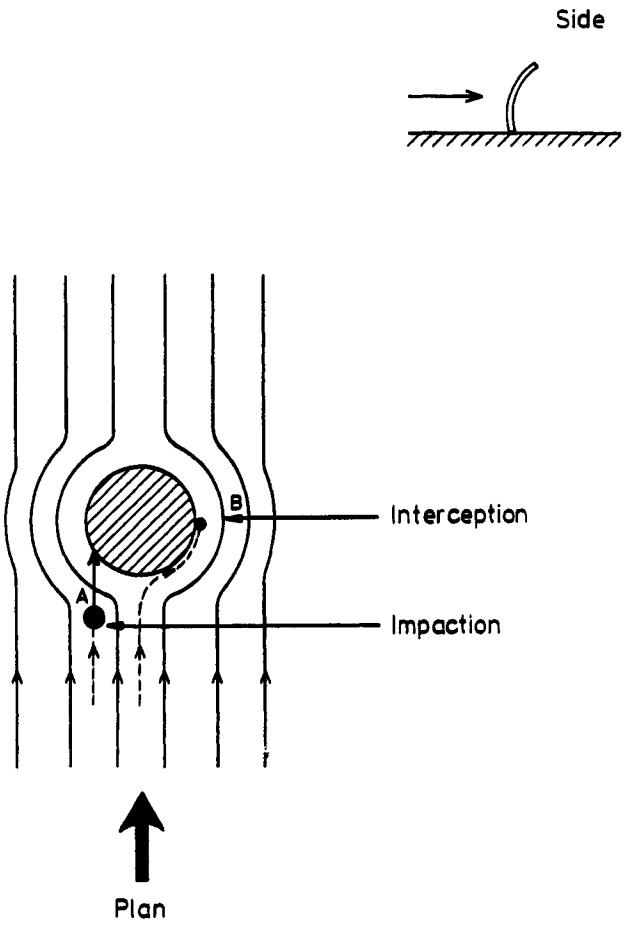

Figure 4.5. Interception and impaction of particles. Impaction is represented by particle A, which crosses the streamlines due to its inertia. The lighter particle, B, follows the streamlines but comes sufficiently close to the surface to make contact.)

the potential importance of wet deposition as a removal process was demonstrated clearly in the aftermath of the Chernobyl nuclear reactor accident, when levels of deposition in the UK were closely related to the amounts of precipitation falling during the passage of the main plume.[77,78]

Wet deposition can be described as resulting from either in-cloud or below-cloud scavenging processes. The importance of each of these processes depends on the release height and the level of the cloud base, but generally in-cloud scavenging tends to dominate with increasing distance from a source.

Measurements of wet deposition often have been described using a scavenging ratio, where:

$$\text{scavenging ratio} = \frac{\text{concentration of material in rain}}{\text{concentration of material in air}} \tag{4.31}$$

The concentrations in Equation 4.31 usually are measured in terms of mass per unit mass, and typical ratios are of the order of several hundred to a thousand or more for atmospheric trace species.[79] It is important to note that some authors express concentrations in terms of mass per unit volume and these result in scavenging ratios that are higher by a factor of approximately 840.

The scavenging ratio is a useful tool for predicting wet deposition levels around a source or receptor region. However, it gives little insight into the factors that affect wet deposition, not least since atmospheric concentration is measured near ground level and this may bear no relation to air concentrations in clouds or at altitude.

It is helpful to consider the processes that result in wet deposition and the formation of precipitation. On most occasions in temperate latitudes, precipitation usually occurs via the Bergeron process. This process describes the formation and growth of ice crystals at the expense of super-cooled water droplets and, hence, precipitation begins as snow. The occurrence of rain or sleet depends on whether the precipitation has melted totally or partially on collection at ground level. Thus, in clouds, ice crystals and super-cooled water droplets coexist with water vapor exchanging between each phase. It is important to note, however, that the Bergeron process does not describe all forms of precipitation formation and there are clear examples in storm systems, especially in tropical latitudes, in which the entire cloud is below freezing level. In these cases, rain is formed by the agglomeration of droplets.

If we consider the formation of in-cloud elements (i.e., ice crystals and water droplets), each is formed by an individual nucleus. There are two categories: the ice nuclei which form ice crystals, and cloud condensation nuclei, which form water droplets. Ice nuclei are relatively scarce, tend to be insoluble and might include particulate materials derived from a soil source. Cloud condensation nuclei include hygroscopic materials and often contain or adsorb pollutants. Although larger particles are the more efficient cloud condensation nuclei, most are smaller than 1 μm diameter.

Concentrations of material in-cloud are much greater in the droplet phase than in the ice phase. Consequently, any droplets that are intercepted by falling ice crystals might contribute considerably to wet deposition. This has been noted when rimed snowflakes (i.e., snowflakes that have frozen droplets associated with them) have been found to contain much greater amounts of pollutants than unrimed ones.[80,81]

In addition to the nucleation processes that occur in-cloud, particulate material can be incorporated into pre-existing droplets. Diffusion and phoretic effects dominate the efficiency of this process, although the relative importance of these processes is poorly understood. In-cloud scavenging experiments are notoriously difficult to undertake and involve the injection of material into clouds via either rockets[82] or aircraft.[83,84]

Below-cloud scavenging occurs due to the collection of material by falling hydrometeors. The processes that result in the collection of atmospheric aerosols can include the effects of Brownian motion, interception and impaction, and there are, consequently, analogies between the mechanisms of below-cloud scavenging and dry deposition (with the exception that sedimentation does not play a significant role in the capture of particles by falling precipitation).

The scavenging coefficient (Λ) often has been used to describe below-cloud scavenging,[85] where

$$\frac{d\chi}{dt} = - \Lambda\chi \tag{4.32}$$

in which χ is the concentration of material available for scavenging. It can be easily shown that

$$\chi_t = \chi_0 \exp(- \Lambda t) \tag{4.33}$$

where χ_t and χ_o are the atmospheric concentrations of material at a time equal to 0 and t. For the simple case of uniformly sized rain droplets, Λ can be related to the removal efficiency of a monodisperse aerosol[86] as

$$\Lambda = NV_T E\pi R^2 , \tag{4.34}$$

in which N is the number of raindrops per unit volume, with a terminal velocity equal to V_t and radius R, which have a collection efficiency (i.e., fraction of material collected in the volume swept out by the falling hydrometer) equal to E. For a range of raindrop sizes, Λ can be given[87] as:

$$\Lambda\ (a,R_s) = \int_0^{oo} \pi\ R^2\ V_t\ (R)\ E\ (a,R)\ N(R)\ dR \tag{4.35}$$

in which a is particle radius, N(R) dR is the number of raindrops per unit volume with radii R to R + dR and R_s refers to a particular raindrop spectrum.

Measurements of below-cloud scavenging have been based to a large extent on laboratory studies,[88] while field measurements have included measurements of the depletion of adventitious atmospheric species[89,90] and the measurement of released materials in collected precipitation.[91,92]

Precipitation intensity is an important parameter when evaluating below-cloud scavenging because both the number and size of raindrops depend on this. May[92] calculated scavenging coefficients for various precipitation intensities using a raindrop spectrum determined by Best.[93] Slinn[94] recommended that the result of Mason,[95] regarding the mean size of drops, could be used to calculate Λ in fairly steady rain, thus eliminating the need for a detailed raindrop size distribution. Here

$$R_m = 3.5 \times 10^{-4} \left(\frac{J}{1\ mm\ h^{-1}}\right)^{0.25} \tag{4.36}$$

where J is the rainfall intensity (mm h^{-1}) and R_m is the mass mean raindrop radius (m). From Equation (4.34), it can be shown that

$$\Lambda \approx \frac{0.75\ J}{R} \tag{4.37}$$

since

$$J = \frac{4}{3}\ \pi\ R^3\ NV_t \tag{4.38}$$

Slinn,[94] using R_m, approximated

$$\Lambda = \frac{c\,JE(a,R_m)}{R_m} \tag{4.39}$$

where c is a constant that may have a value around 0.5 for frontal rain.[96]

For typical precipitation intensities of 1 to 2 mm h^{-1}, it may be anticipated that values of Λ, for particles of several micrometers diameter, will be in the range 10^{-4} to 10^{-3} s^{-1}. However, there are few supportive data and the apparent observation, in some cases,[97] of collection efficiencies greater than 100% have led to increased uncertainty. There are even greater uncertainties in appropriate values of Λ to be used for <1 μm diameter particles.

Some predictive models for below-cloud scavenging adopt values of Λ/J. These are often assumed to be a constant [typically 10^{-4} s^{-1} (mm $h^{-1})^{-1}$], although there are few supportive experimental data for such an assumption.

As previously noted, the deposition of fog or cloud droplets is a potentially important process.[98,99] The formation of the droplets will include some of the nucleation processes that result in wet deposition. However, the droplets are sufficiently small to be unaffected by gravity and the deposition of them can be described as being similar to the dry deposition of large particles. The importance of such deposition, in relation to dry and wet deposition, is strongly dependent on location. However, the potential contribution of deposition in fog and cloud droplets has been noted widely, not least because the concentration of trace species has been found to be high in the droplets.[100]

THE RESUSPENSION OF MATERIAL INTO THE ATMOSPHERE

The terms resuspension and re-entrainment are used generally to describe the processes by which any deposited material might become airborne. Strictly, only aerially deposited material can become resuspended, although the term often is used also to describe the suspension of material from spills, etc.

Early interest in entrainment was due to the effects of erosion and soil transport.[101,102] However, with the advent of nuclear technology, the resuspension of radionuclides has received wide attention.[103,104] Other interests in resuspension include the fate of industrial spills,[105] the transport of chlorinated hydrocarbon pesticides,[106] the spreading of crop disease[107] and the transmission of human disease.[108]

The interests in resuspension are two-fold. First, an inhalation hazard might occur and, second, there can be a resulting spread of contamination due to the deposition of resuspended material. It is important also to note that an inhalation hazard occurs primarily due to small particles, although it is large particles that are dominant in the spread of contami-

nation. Furthermore, resuspended material often is associated with surface dust or soil particles and the particle size range of resuspended material may bear no resemblance to the particle size range of the depositing species.[109]

Because the inhalation of resuspended material has been of concern, the resuspension factor (RF) has been used to describe resuspension.

$$RF(m^{-1}) = \frac{\text{atmospheric concentration}}{\text{ground contamination}} \tag{4.40}$$

There have been difficulties in ascribing the height of the concentration measurement and the depth of surface material that should be sampled to determine ground contamination. In addition, there is an underlying assumption, in the use of the resuspension factor, that airborne material originates from the immediately underlying soil and that downwind transport need not be considered. This is unlikely to be the case, although the resuspension factor still can be used in inhalation dose assessments for areas of fairly uniform contamination.

An alternative to the resuspension factor is the resuspension rate, R_r, where

$$R_r = \frac{\text{resuspension flux}}{\text{ground contamination}} \tag{4.41}$$

The use of a resuspension rate, rather than a resuspension factor, has been supported because this can be incorporated into a diffusion model to enable the prediction of downwind air concentrations.[110] However, in practice it is difficult to measure. The use of it suffers the same disadvantages as that of the resuspension factor, with regard to determining ground contamination.

The availability of material for resuspension declines with time lapsed after deposition. This decline has been expressed in terms of a negative exponential function[111] and as an inverse relationship.[112] Reeks et al.[113] have considered the theoretical aspects of resuspension, where surface particles are resident in potential energy wells and are periodically resuspended by the penetration of turbulent eddies through the laminar sub-layer. They concluded that resuspension was inversely proportional to time and there has been further supporting evidence of this relationship in describing resuspension from grass and concrete surfaces in wind tunnel experiments.[114] It is important to note that the decline in resuspension with time is not due to a depletion in the amount of deposited material, but is due to material becoming less erodible.[115] Consequently, the time after the initial deposition episode is of fundamental importance in the interpretation of resuspension data, and variations of this might go some way toward explaining the extremely large range of resuspension results.[8]

Resuspension rate has been found by several authors to increase in proportion to the wind speed raised to a power,[116] although the exact values of this power have varied, being typically in the range 1 to 6, with the possibility of the power being time dependent.[117] Nevertheless, such relationships are strong evidence of the episodic nature of wind-generated resuspension.

A possible important factor in the resuspension process is the occurrence of saltating particles[118] (i.e., particles that leave the surface, but that have such high sedimentation velocities that they do not remain suspended). Saltating material has been found to be of great importance in erosion processes and there has been a threshold velocity found for