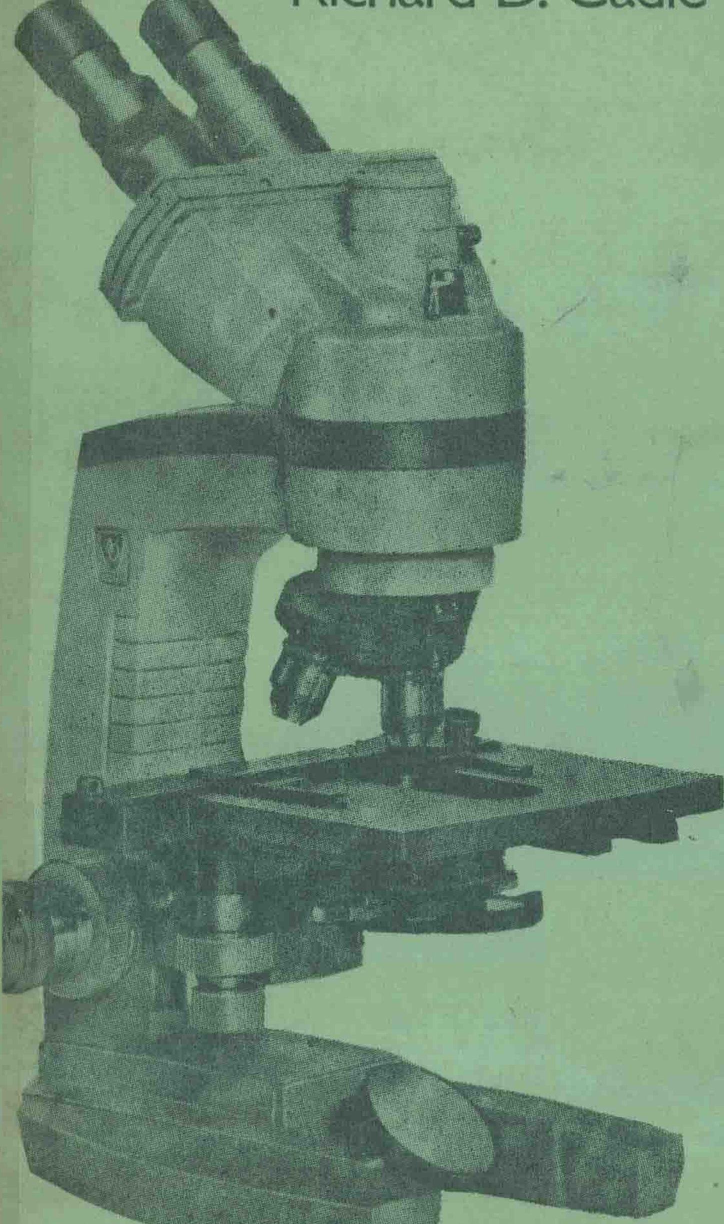
THE MEASUREMENT OF AIRBORNE PARTICLES

Richard D. Cadle



THE MEASUREMENT OF AIRBORNE PARTICLES

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（内部交流）

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SERIES PREFACE

Environmental Science and Technology

The Environmental Science and Technology Series of Monographs, Textbooks, and Advances is devoted to the study of the quality of the environment and to the technology of its conservation. Environmental science therefore relates to the chemical, physical, and biological changes in the environment through contamination or modification, to the physical nature and biological behavior of air, water, soil, food, and waste as they are affected by man's agricultural, industrial, and social activities, and to the application of science and technology to the control and improvement of environmental quality.

The deterioration of environmental quality, which began when man first collected into villages and utilized fire, has existed as a serious problem since the industrial revolution. In the last half of the twentieth century, under the ever-increasing impacts of exponentially increasing population and of industrializing society, environmental contamination of air, water, soil, and food has become a threat to the continued existence of many plant and animal communities of the ecosystem and may ultimately threaten the very survival of the human race.

It seems clear that if we are to preserve for future generations some semblance of the biological order of the world of the past and hope to improve on the deteriorating standards of urban public health, environmental science and technology must quickly come to play a dominant role in designing our social and industrial structure for tomorrow. Scientifically rigorous criteria of environmental quality must be developed. Based in part on these criteria, realistic standards must be established and our technological progress must be tailored to meet them. It is obvious that civilization will continue to require increasing amounts of fuel, transportation, industrial chemicals, fertilizers, pesticides, and countless other products and that it will continue to produce waste products of all descriptions. What is urgently needed is a total systems approach to modern civilization through which the pooled talents of scientists and engineers, in cooperation with social scientists and the medical profession, can be focused on the development of order and equilibrium to the presently disparate segments of the human environment. Most of the skills and tools that are needed are already in existence. Surely a technology that has created such manifold environmental problems is also capable of solving them. It is our hope that this Series in Environmental Sciences and Technology will not only serve to make this challenge more explicit to the established professional but that it also will help to stimulate the student toward the career opportunities in this vital area.

Robert L. Metcalf
James N. Pitts, Jr.
Werner Stumm

PREFACE

The importance of airborne particles to humanity must be obvious to most persons; it hardly needs to be emphasized here. For example, we all live in a great aerosol system, our atmosphere, and the particles it contains have vast influences on our climate and on our health. Much of our technology involves the production, use, and often the elimination of airborne particles. Growing concern about man's impact on the quality of our environment, and specifically about the increasing burden of atmospheric particles for which man is responsible on local, regional, and even global scales, has accelerated investigations of airborne particles by colleges, universities, government agencies, and industrial laboratories. Measurement of the size distributions, shapes, and concentrations of the particles is essential to such investigations; the techniques and equipment for making such measurements is the subject of this book.

The proposition that the development and expansion of technology will occur to fill a need is axiomatic, and the art and science of aerosol particle measurement is no exception. In 1955 I wrote a small book entitled *Particle Size Determination* which was the first monograph devoted to this subject. Particles in various media such as powders, pastes, suspensions in liquids, and suspensions in air were considered. Since then a number of excellent books on particle measurement have been written that have enabled the reader to keep up to date in this rapidly expanding field. Now both the theory and art of airborne particle measurement is so extensive that the treatment of the subject in depth nearly requires the publication of a monograph dealing with that subject alone. I have undertaken the task of writing such a book because I have directed and carried out laboratory and field investigations of aerosols ever since being asked to participate in a study of Los Angeles smog many years ago. Some of the field investigations have become adventures when they involved determining the contributions to atmospheric trace constituents of the rain forests of Panama and Amazonia, volcanoes in various parts of the world, and nuclear explosions.

This book has been written primarily as an aid to the scientist or engineer faced with the problem of making aerosol particle measurements. However, it should also be useful to a person desiring to learn more about this field, and as a textbook or source of supplementary material for college or university courses concerned with aerosol science and technology. The book deals to a greater extent than most monographs with commercially developed and marketed instruments. I believe that this is essential, since many of the greatest improvements made in recent years in the particle measurement art have occurred in industrial laboratories. Furthermore, reference to specific manufacturers should aid the reader in locating needed equipment. Clearly, not all commercial sources of airborne particle measuring equipment could be included, and the omission of any such source should not be interpreted to be derogatory. Furthermore, the selection of illustrations of commercial equipment in most cases depended on their being supplied by the manufacturer.

The first chapter deals to a considerable extent with particle statistics and data presentation, emphasizing recent developments. The organization of the chapters dealing with actual measurement techniques is fairly straightforward, each chapter discussing first theory and then methodology. However, the organization of the material on impaction required some soul searching on my part, because impaction techniques can be used both to collect and to classify particles according to size. Impaction techniques designed merely to collect particles are described in Chapter 2, and multistage impactors are discussed in Chapter 5. I hope that this arrangement is not too confusing. The last chapter deals with the important subject of isokinetic sampling.

More space has been devoted to filtration and to optical microscopy than to many of the other subjects. This is partially because filtration and optical microscopy are very old and very useful techniques that have received a great deal of attention. Furthermore, they are available to most scientists and engineers with the outlay of little or no money. Another reason for discussing optical microscopy in great detail is that although microscopes are part of the equipment of a great many laboratories, few persons know how to use them to best advantage.

I am pleased to acknowledge the help of many persons in the preparation of this volume. I especially wish to thank Ms. Nadine Perkey who

coped so successfully with my nearly illegible handwriting while typing the manuscript, Dr. J. P. Lodge, Jr., for his helpful scientific and technical comments, and Ms. Kay Redman who helped polish the text.

R. D. CADLE

Boulder, Colorado
January, 1975

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INTRODUCTION

Aerosols can be defined as any relatively stable suspension of particles in a gas, especially in air, including both the continuous (gaseous) and discontinuous (particulate) phases. Since World War II, especially during the last decade, great advances have been made in our knowledge of the behavior of aerosols and in the techniques available for studying them. Perhaps the greatest incentive for the present accelerated research on aerosols has been the increasing concern about the impact of man on his environment, particularly the extent to which he is polluting the atmosphere—locally, regionally, and worldwide. Another incentive for this research is the need in many modern devices for closer tolerances that can be achieved only by manufacturing them in relatively dust-free rooms, the so-called clean rooms. Studies of airborne particles are an important aspect of industrial hygiene, weather modification, and studies of the natural atmosphere. Furthermore, investigations of the production, characterization, and behavior of aerosols has become a scholarly discipline.

Often, the most important part of any aerosol investigation is measuring the suspended particles. Here, measurement refers to the determination of size distributions, concentrations, and shapes. Such measurements, of course, constitute one aspect of fine-particle technology, and methods for making them have been described in this context in many publications.[1-4] During recent years, a proliferation of techniques, especially automatic methods, has developed for measuring the particles in various environments (powders, liquid suspensions, and aerosols, for example); and the measurement of airborne particles is now a specialty. In fact, many of the methods developed for the measurement of aerosol particles are applicable only to particles suspended in a gas.

1. SOME DEFINITIONS

1.1. Particles

The term "particle" can be defined as any object having precise physical boundaries in all directions, and the meaning of the term "individual particles" in an aerosol is often obvious. However, when the suspended particles are aggregates, when there are particles of different types such as of different chemical composition, or when the size distributions are very wide or otherwise complex, the investigator must be careful to precisely define the particles with which he is concerned. Then the methods selected for making the desired measurement must be consistent with that definition. If the suspension contains aggregates, the investigator must decide whether he should measure the aggregates, the individual particles in the aggregates, or both. A common error may occur when an aerosol contains two or more types of particles, each type having a greatly different size distribution. If the investigator is interested in only one of the particle types and is not aware of the presence of the others, gross measurement errors may result.

Errors from this source can arise, for example, when an aerosol is prepared with an aspirator by dispersing a dilute suspension of particles in a volatile liquid. Many of the droplets produced contain the insoluble particles, and evaporation of the volatile liquid leaves the particles in suspension in air. However, if the liquid contained traces of dissolved, nonvolatile material, such as a wetting agent, evaporation of the droplets will produce an aerosol containing, in addition to the desired particles, large numbers of very small particles consisting only of the nonvolatile material. Even if only one type of particle is in suspension, if the size distribution is very wide, a decision often must be made with regard to the size range of interest and the appropriate measurement technique to be employed.

1.2. Particle Size

Particle size and particle shape also need precise definition if they are to be measured. The need to define particle shape if it is to be described quantitatively is usually apparent, but the similar need to define particle size is often overlooked. One of the most common and frustrating omissions in the scientific and technical literature dealing with fine particles is the failure to state whether particle "size" refers to radii or diameters. But, even if this is indicated, the term diameter (or radius) is unambiguous only for spheres. When all the particles in a system such as an aerosol have the same shape (such as cubic), then the problem of defining diameter in terms of the dimensions of the particles is relatively easy. However, when the particles vary markedly in shape, definition is much more difficult.

Diameters of irregular particles can be defined in terms of the geometry of the individual particles or in terms of their physical properties. Diameters of the former type are often described as statistical because they have much meaning only for averages of the measurements of a large number of irregular particles. Sieving as a method of size determination (which is not discussed in this book because it is seldom if ever appropriate for aerosol particles) can be considered to yield statistical diameters, since the results depend directly on the dimensions of the particles and are averaged by the method for a large number of particles.

Diameters defined in terms of physical properties are determined by measuring properties such as light scattering, sedimentation rates, or rates of Brownian diffusion.

Perhaps the most widely used statistical diameter is the diameter of a circle whose area is the same as that of the area of the particle projected onto a surface. It was originally used primarily for particle measurement by optical microscopy, but it is being used more and more in connection with automatic and semiautomatic sizing methods.

This definition is easy to apply when the particles are regular in shape, but it is almost impossible to apply to certain aggregates. An example is the particles shown in the electron micrograph of Figure 1-1. They were collected with impactors mounted on U.S. Air Force RB-57F aircraft from the stratosphere at about 18 km altitude a few weeks after extentensive brush fires in California, and they may have been particles of wood smoke.[5] One way to treat such aggregates is to determine the sizes of the individual ("ultimate") particles and the number of such particles in each aggregate measured.

Another difficulty often arises when airborne droplets are collected on hard surfaces, for example, by impaction. The droplets tend to spread on the surface and may, upon pulling together, leave satellite droplets. The resulting pattern even may provide a means for identifying the major constituent of the droplets.[6] Figure 1-2 is an electron micrograph of particles collected from the Antarctic atmosphere near McMurdo Sound;[7]

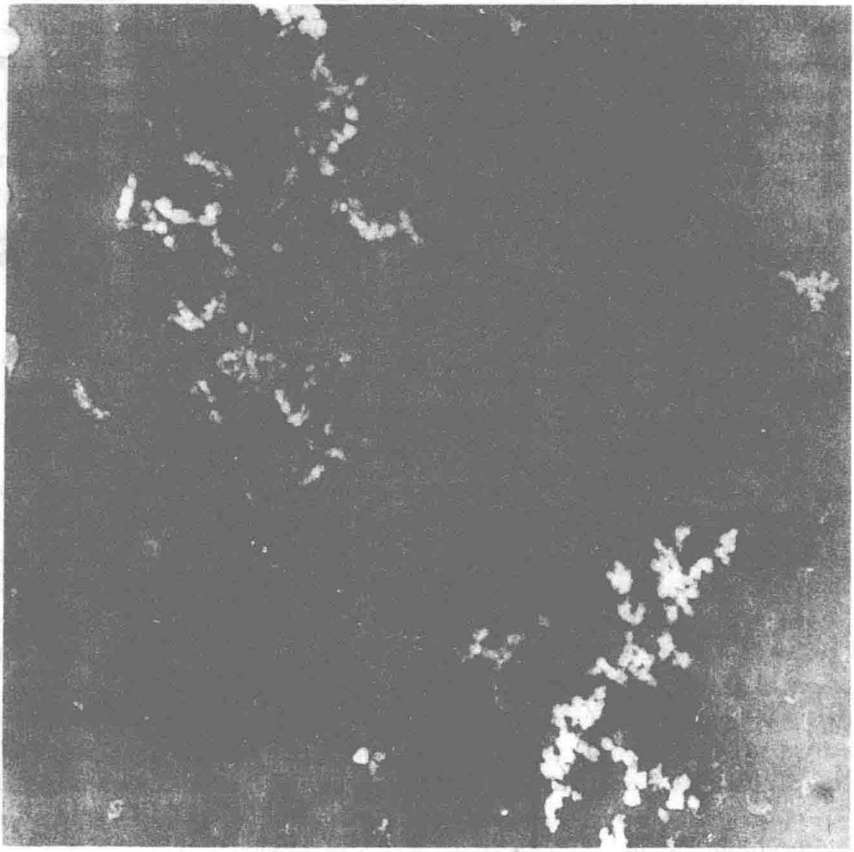

Figure 1-1. Particles collected by impaction from the midlatitude stratosphere at 18 km altitude on 21 October 1970. The distance across the electron micrograph is 6 μm. From Cadle, R. D., EOS, Transactions, American Geophysical Union, 53, 812 (1972). Copyright by the American Geophysical Union. Magnification 19,314×.

Figure 1-3 is an electron micrograph of particles collected from the fume from the lava fountains of Kilauea volcano in Hawaii in 1967.[8] The patterns are typical of sulfuric acid droplets, a common atmospheric constituent. If size distributions are to be determined from measurements of the images of the collected droplets, attempts must be made to estimate the original droplet volume. Some methods for doing this are discussed in the section on microscopy.

Other definitions of diameter are based on the assumption that the length (l), breadth, (b), and thickness (t) of the particle can be measured.

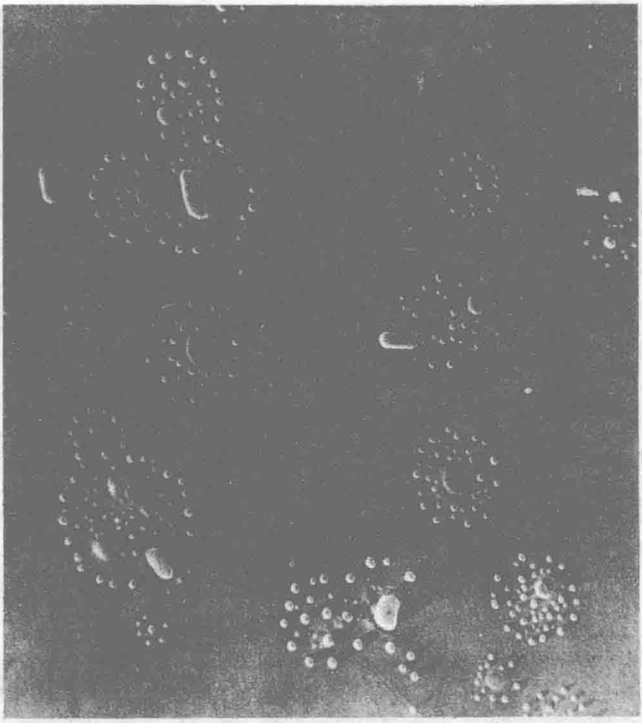

Figure 1-2. Electron micrograph of particles impacted from Antarctic air.[7] Field of view is 6 μm across. The particles were probably impure sulfuric acid.

These can be presented separately or the diameter can be defined as the arithmetical average of the three dimensions

$$d = \left(\frac{1}{3}\right)(l + b + t) \tag{1-1}$$

The dimensions also can be used to define the diameter as the length of the side of a cube of equal volume

$$l = \sqrt[3]{lbt} \tag{1-2}$$

Martin[9] proposed the following definition of diameter. It is the distance between opposite sides of the particle, measured crosswise of the particle, and on a line bisecting the projected area (Figure 1-4). The diameters must always be measured in the same direction if the results are to be statistically significant. For example, if the measurements are

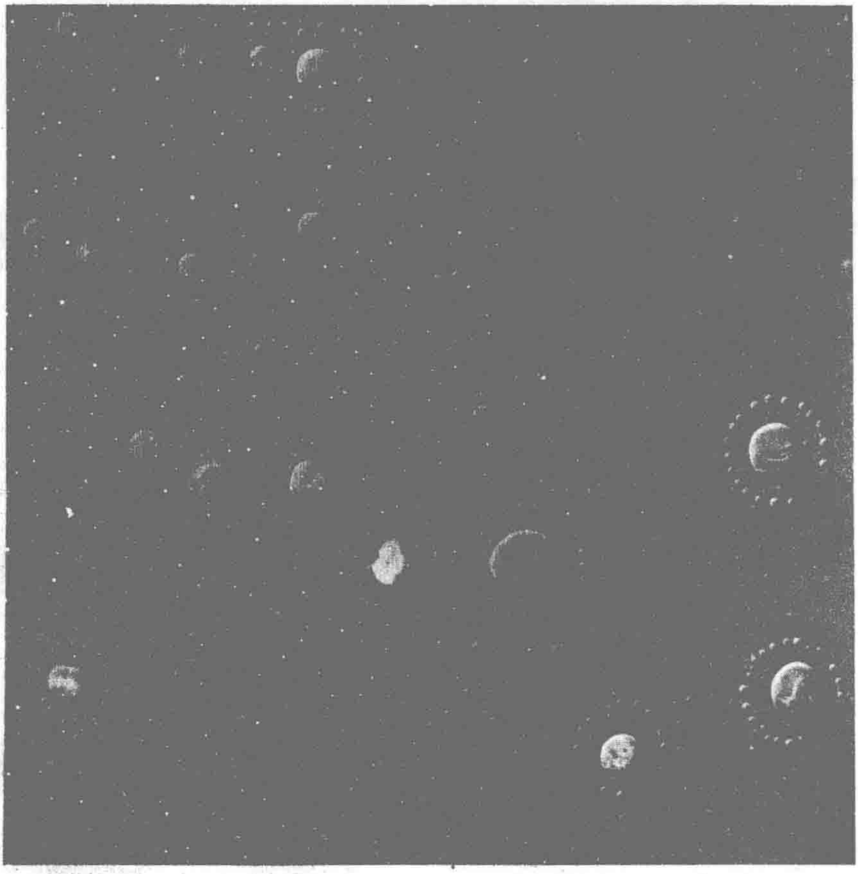

Figure 1-3. Electron micrograph of particles collected by impaction from the fume from the lava fountains of Halemaumau crater of Kilauea volcano during the eruption of 1967–1968. The particles consisted of impure sulfuric acid. Distance across micrograph is 25 μm^8.

made with a microscope, the direction parallel to the bottom of the field is convenient. Tomkeiff[10] and Moran[11] have suggested that the relationship between Martin's diameter and the area per unit volume (specific surface) of a powder can be calculated from the expression

$$\text{Martin's diameter} = \frac{4}{D_p S_v}$$

where D_p is the packing density and S_v is the specific surface.

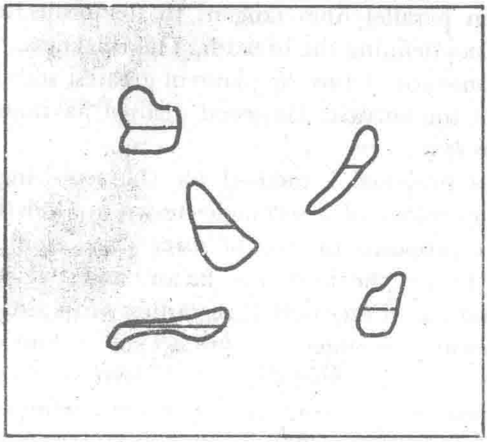

Figure 1-4. Martin's diameters.

Feret's diameter[12] is the distance between two tangents on opposite sides of the particle. As in the case of Martin's diameter, the tangents must be parallel to an arbitrarily fixed direction.

Elongated particles such as fibers may be described merely by length, by thickness, or by both.[13]

1.3. Particle Shape

Defining particle shape is often even more difficult than defining particle size.

Two general approaches to determining and reporting the shapes of particles are based on (1) measuring the dimensions of individual particles, and (2) estimating the mean geometric properties from two different properties of the aerosol, which are influenced to different extents by the shape of the particles. The first approach has already been implied; that is, measuring the length, breadth, and thickness of a large number of the collected particles. Heywood[14] has discussed this approach to determining particle shape and the effect of shape on the results of various types of particle size determinations. He proposed that the particle be assumed to be resting on a plane in the position of greatest stability. The breadth, b, he defined as the distance between two parallel lines tangent to the projection of the particle on the plane and placed so that the distance between them is as small as possible. The length, l, is the distance between parallel lines tangent to the projection and perpendicular to the lines defining the breadth. The thickness, t, is the distance between two planes parallel to the plane of greatest stability and tangent to the surface of the particle. Heywood defined flakiness, F, as b/t and elongation, E, as l/b.

Hausner[15] has proposed a method for characterizing particle shape based on the dimensions of a rectangle drawn around the projection of the particle. He proposed the use of three dimensionless ratios, called the elongation factor, the bulkiness factor, and the surface factor to characterize the shape of the particle regardless of its size. This, of course, is a two-dimensional treatment, as are several definitions of diameter described above but, to be precise, the definition of shape and diameter is a two-dimensional treatment, as are several definitions of diameter plate- or flake-type particles. Hausner suggested that the rectangle of minimum area drawn around the particle projection be used. The elongation of the particle is given by

$$x = a'/b' \tag{1-3}$$

where x is the elongation factor, and a' and b' are the side lengths of the rectangle, a' being the longer. Hausner also defines a bulkiness factor

$$y = \frac{A}{a'xb'} \tag{1-4}$$

where y is the bulkiness factor and A is the projected area of the particle. Thus, y is unity for a particle that just fills the rectangle but is usually much less than this.

Since, for particles of identical shapes, the ratio of surface area to volume decreases with increasing particle size, the rectangle dimensions can-

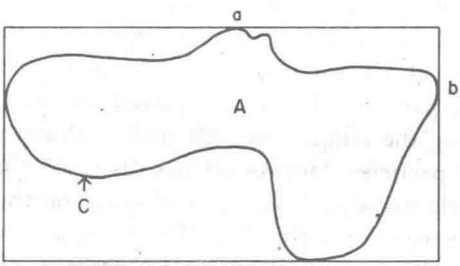

Figure 1-5. Characterization of particle shape using the method of Hausner;[15] a and b are the lengths of the sides of the rectangle, A is the projected particle area and C is the circumference.