MICROPHYSICS OF CLOUDS AND PRECIPITATION

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MICROPHYSICS OF CLOUDS AND PRECIPITATION

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Second revised and expanded edition
with an introduction to
cloud chemistry and cloud electricity

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PREFACE TO THE FIRST EDITION

Cloud physics has achieved such a voluminous literature over the past few decades that a significant quantitative study of the entire field would prove unwieldy. This book concentrates on one major aspect: *cloud microphysics*, which involves the processes that lead to the formation of individual cloud and precipitation particles.

Common practice has shown that one may distinguish among the following additional major aspects: *cloud dynamics*, which is concerned with the physics responsible for the macroscopic features of clouds; *cloud electricity*, which deals with the electrical structure of clouds and the electrification processes of cloud and precipitation particles; and *cloud optics* and *radar meteorology*, which describe the effects of electromagnetic waves interacting with clouds and precipitation. Another field intimately related to cloud physics is *atmospheric chemistry*, which involves the chemical composition of the atmosphere and the life cycle and characteristics of its gaseous and particulate constituents.

In view of the natural interdependence of the various aspects of cloud physics, the subject of microphysics cannot be discussed very meaningfully out of context. Therefore, we have found it necessary to touch briefly upon a few simple and basic concepts of cloud dynamics and thermodynamics, and to provide an account of the major characteristics of atmospheric aerosol particles. We have also included a separate chapter on some of the effects of electric fields and charges on the precipitation-forming processes.

The present book grew out of a series of lectures given to upper division undergraduate and graduate students at the Department of Atmospheric Sciences of the University of California at Los Angeles (UCLA), and at the Department of Physics of the New Mexico Institute of Mining and Technology at Socorro (New Mexico Tech.). We have made no attempt to be complete in a historical sense, nor to account for all the work which has appeared in the literature on cloud microphysics. Since the subject matter involves a multitude of phenomena from numerous branches of physical science, it is impossible to make such a book truly self-contained. Nevertheless, we have considered it worthwhile to go as far as possible in that direction, hoping thereby to enhance the logical structure and usefulness of the work. In keeping with this goal, our emphasis has been on the basic concepts of the field.

This book is directed primarily to upper division and graduate level students who are interested in cloud physics or aerosol physics. Since no specialized knowledge in meteorology or any other geophysical science is presumed, the material presented should be accessible to any student of physical science who has had the more or less usual undergraduate bill of fare which includes a general background in physics, physical chemistry, and mathematics. We also hope the book will be of value to those engaged in relevant areas of teaching and research; also, we hope it will provide a source of useful information for professionals working in related fields, such as air chemistry, air pollution, and weather modification.

In the preparation of this book we have incurred many debts. One of us (H.R.P.) is extremely grateful to his long time associate Prof. A. E. Hamielec of McMaster University at Hamilton, Canada, whose generous support provided the basis for solving many of the hydrodynamic problems reported in this book. Gratitude is also gladly expressed to the faculty and research associates at the Meteorological Institute of the Johannes Gutenberg University of Mainz and at the Max Planck Institute for Chemistry at Mainz, in particular to Profs. K. Bullrich and C. Junge, and Drs. G. Hänel, F. Herbert, R. Jaenicke, and P. Winkler for the assistance received during two stays at Mainz while on sabbatical leave from UCLA. In addition, sincere thanks are extended to the Alexander von Humboldt Foundation for a U.S. Senior Scientist Award which made possible the second extended visit at Mainz. Also, one of us (J.D.K.) is grateful to Drs. C. S. Chiu, P. C. Chen, and D. T. Gillespie for informative discussions, and to Prof. M. Brook and Dr. S. Barr for providing time away from other duties. Appreciation is expressed also to the National Center for Atmospheric Research (NCAR) for the assistance provided during a summer visit.

A large number of figures and tables presented in this book have been adapted from the literature. The publishers involved have been most considerate in granting us the rights for this adaptation. In all cases, references to sources are made in the captions.

Our own research reported in this book has been supported over the years by the U.S. National Science Foundation. We would like to acknowledge not only this support, but also the courteous, informal, and understanding manner in which the Foundation's Officers, Drs. F. White, P. Wyckoff, E. Bierly, and F. Eden, conducted their official business with us.

Special thanks go also to the editors of the D. Reidel Publishing Company of Dordrecht, Holland, for providing a fruitful relationship with us.

Finally, we wish to express our sincere appreciation for the invaluable assistance of T. Feliciello, A. C. Rizos, and P. Sanders, who typed the manuscript, and to B. J. Gladstone who drew all the diagrams.

Los Angeles, H. R. PRUPPACHER
March 1978
Los Alamos, J. D. KLETT

PREFACE TO THE SECOND EDITION

In the intervening eighteen years since the appearance of the first edition, research in cloud microphysics has continued to expand at a rapid rate. In fact, we have found it necessary to consider for inclusion in this edition the contents of over 5,000 articles, as well as dozens of books and conference proceedings published since the first edition. Our approach to assimilating this material follows the philosophy of the first edition, namely to attempt a balance between providing a necessary body of descriptive and empirical knowledge, and a framework of theoretical generalities and principles with which to rationalize the otherwise unmanageable mountain of experimental facts. Such an effort naturally entails compromises and personal choices, as a truly exhaustive and completely coherent account of a subject this large cannot be confined within the bounds of a single volume of an acceptable length. Nevertheless, we feel that the present volume does accommodate the most significant advances that have occurred, and that it has been possible to close some of the gaps and answer some of the major questions which characterized the incompleteness of the subject at the time of the first edition.

As before, we have again attempted to enhance the appeal and clarity of the book by making it as self-contained as possible. Our success in this respect has been limited, not only because of the sheer volume of material, but also because of a shift in style of the theoretical approach to the subject. Now that nearly everyone has access to inexpensive desktop computers with more power than mainframes at the time of the first edition, and similar access to greatly improved and easily implemented numerical modeling software, a tendency has developed to address theoretical issues by constructing and then incrementally augmenting numerical models of great complexity, often of an ad hoc nature and with many adjustable parameters. The underlying assumption that more and more physics can successfully be encoded this way into larger and larger programs is sometimes subject to challenge; in any case, the resulting algorithms are often so complex that they and their results have to be accepted largely on faith by other researchers. It is obviously difficult to include an account of such theoretical work in a way that is truly self-contained and logically complete.

We have also had to continue to be extremely restrictive in treating fields intimately related to cloud microphysics. Thus, as in our first edition, we could touch only briefly on some simple concepts of cloud dynamics, and refer in places only to the results of cloud dynamic models which include detailed microphysics. (An excellent text on cloud dynamics is now available in the treatise by Cotton and Anthes (1989).) We also had to leave out the extensive field of the interaction between clouds and electromagnetic radiation, although we sometimes refer to results derived from radar cloud studies and from studies on the effects of solar and terrestrial radiation on the microstructure of clouds. Also, in the chapter on cloud electricity we have had to omit many facets of clear weather electricity, and the subject of the physics of lightning. On the other hand, we have amplified the present edition by the inclusion of a chapter on cloud chemistry (Chapter 17). This was prompted by the seriousness with which worldwide ecological problems related to air, water, and ground pollution are viewed by the scientific community in general. Our treatment of the subject is restricted to some basic processes that must be considered in current pollution transport models.

Other changes in the book worth noting here include: (1) The descriptive material in Chapter 2 on the microstructural features of clouds has been updated and includes more diagrams to assist modelers, and much more information on cirrus clouds. (2) The section on the structure of water in Chapter 3 reflects our greatly improved knowledge of the specific heat, latent heat, and other properties of water, all the way down to −40°C; this supersedes previous extrapolations from the Smithsonian Tables. As an example, the new data on the activation energy for molecular transfer at the ice-water interface leads to homogeneous ice nucleation rates in much better agreement with cirrus observations (described in Chapter 7). Also, a distinct statistical mechanics theory for ice nucleation is now included, and it is shown that the thermodynamic data are consistent with the molecular data from ice physics research. (3) In Chapter 5 the values for surface tension and interface energy below 0°C are recomputed due to the new results in Chapter 3. (4) Size distribution measurements of the atmospheric aerosol now extend down to 0.01 μm and lower. This new data, and enhanced discussions of gas-to-particle and drop-to-particle conversion, are included in Chapter 8. Also included is new information on aerosols over the North and South Polar regions, which is of relevance to the phenomenon of the Ozone Hole. (5) In Chapter 9, new statistical mechanics modeling results for the heterogeneous nucleation of ice on silver iodide and silicates supplement the previous thermodynamic approach. (6) Numerical simulations of flow about spheres at Reynolds numbers too high for steady axisymmetric flow, as well as for flow past cylinders and three types of snow crystal shapes, are now included in Chapter 10. This gives rise to new ventilation coefficients, hydrodynamic drag, and terminal velocities. Also, new data and modeling concerning drop breakup and oscillations are provided. As an application, improved non-equilibrium descriptions of oscillating drop shapes are given. (6) An amplified treatment of drop condensation growth in stratus clouds and fogs is given in Chapter 13. The chapter also includes new results on ventilation, and some sensitivity studies on the effects of drop collision and coalescence on the early stages of evolving spectra of cloud drops. (7) New parameterizations of experimental work on drop coalescence are given in Chapter 14. Also, the new flow fields described in Chapter 10 are used to determine collision cross-sections between various combinations of drops, finite-length cylinders, plates, and some other crystal shapes. The problem of turbulence is also revisited, including its effect on the orientation distribution of particles. (8) More complete simulations of stochastic drop breakup and growth are given in Chapter 15, along with an expanded treatment of the method of moments. (9) Chapter 16 has been enlarged with respect to parameterizations of experimental data on graupel, rime, hailstones, and the polycrystallinity of frozen drops; there are also new theoretical modeling results on the growth rate of graupel, snow crystals, and hailstones in dry and wet regimes, on the evolution of ice particle size distributions taking various interactions into account, and on the melting of ice particles. (10) In Chapter 18, the description of strongly electrified clouds and cloud particles based on field studies has been updated and expanded considerably. The major cloud charging mechanisms are reviewed in light of new experimental data, and it is concluded that certain non-inductive mechanisms are dominant and primarily responsible for the tripolar thundercloud charge distribution often observed. The sections on the effects of electric fields and charges on drop shape and disruption, corona discharge, and the enhancement of collection and scavenging processes for various types of cloud particles have also been expanded and improved.

The overall scope and intended audience of the book remain unchanged. In particular, we hope it may provide for the upper division and graduate level student a quantitative survey of cloud microphysics, and that it will be a source of useful information for those engaged in related areas of teaching and research, including the fields of aerosol physics, cloud dynamics, climate modeling, air chemistry, air pollution, and weather modification.

In the preparation of this book, we have again incurred many debts. One of us (H.R.P.) is indebted to the German National Science Foundation (DFG) and to the German Ministry for Research and Technology (BMFT) for generously supporting the research carried out at the Institute for Physics of the Atmosphere at the Johannes-Gutenberg University of Mainz. He is particularly very grateful to his longtime associates Prof. Andrea Flossmann and Dr. Subir Mitra, and to Drs. Stefan Borrmann and Andreas Bott for fruitful and constructive criticism in preparing this text. He is thankful to them, as well as to Profs. K. V. Beard and P. K. Wang, for providing him with original figures, tables, and texts, in part still unpublished. Thanks go also to all his students, who have provided through their questions insights for clarifying many of the concepts presented in this book. He also warmly thanks his wife Monica and his son Lukas for bearing the many years of hardships with him. The second author (J.D.K.) would like to thank Prof. P. Chylek for many stimulating and wide ranging discussions, and Dr. R. A. Sutherland of the Army Research Laboratory of White Sands Missile Range, Dr. M. Farmer of Correa Enterprises Inc., and Mr. J. Serna of the Physical Sciences Laboratory of New Mexico State University for their indirect roles in providing financial support that facilitated the completion of this project. He also thanks his wife Cathy, and Mark, Lindsay, and Ali for keeping it all in perspective with love and good cheer. Finally, we wish to express our sincere gratitude to Mrs. K. Franke who typed the original manuscript, to Mr. Michael Lang for coordinating the text into LaTeX, to Mrs. Cornelia Schrörs who set the tables and the figure captions, to Mr. Werner Klaus Zangi who drew all the diagrams, and to Mrs. Renate Graf-Gries and Andrea Richter who were kind enough to coordinate much of the clerical work.

Again, a number of figures and tables in this book have been adapted from the literature. The publishers and authors involved have been most considerate in granting us the rights for this adaptation. In all cases, references to sources are made in the figure and table captions.

Special thanks also go to the editors of Kluwer Academic Publishers, Dordrecht, in particular to Dr. Mariette de Jong, for providing a fruitful relationship with us.

Mainz, H. R. PRUPPACHER
March 1996
Las Cruces, J. D. KLETT

Hath the rain a father? or who has begotten
the drops of dew?
Out of whose womb came the ice? and the hoary
frost of heaven who has gendered it?
Canst thou lift up thy voice to the clouds,
that abundance of waters may cover thee?
Who can number the clouds in wisdom?
or who can stay the bottles of heaven?
Canst thou send lightnings, that they may go,
and say unto thee, here we are?
Knowest thou the ordinances of heaven? canst
thou set the dominion thereof in the earth?
Job **38**, 28-29, 34, 37, 35, 33

CHAPTER 1

HISTORICAL REVIEW

As one studies the meteorological literature, it soon becomes evident that cloud microphysics is a very young science. In fact, most of the quantitative information on clouds and precipitation, and the processes which are involved in producing them, has been obtained since 1940. Nevertheless, the roots of our present knowledge can be traced back much further. Although a complete account of the development of cloud physics is not available, a wealth of information on the history of meteorology in general can be found in the texts of Körber (1987), Frisinger (1977), Middleton (1965), Khrgian (1959) and Schneider-Carius (1955). Based on these and other sources, we shall sketch here some of the more important events in the history of cloud physics. In so doing we shall be primarily concerned with developments between the 17th century and the 1940's, since ideas prior to that time were based more on speculation and philosophical concepts than scientific fact and principles. As our scope here is almost exclusively restricted to west European and American contributions, we emphasize that no claims for completeness are made.

It was apparently not until the 18th century that efforts were underway to give names to the characteristic forms of clouds. Lamarck (1744-1829), who realized that the forms of clouds are not a matter of chance, was probably the first to formulate a simple cloud classification (1802); however, his efforts received little attention during his lifetime. Howard (1772-1864), who lived almost contemporaneously with Lamarck, published a cloud classification (1803) which, in striking contrast to Lamarck's, was well received and became the basis of the present classification. Hildebrandson (1838-1925) was the first to use photography in the study and classification of cloud forms (1879), and may be regarded as the first to introduce the idea of a cloud atlas. This idea was beautifully realized much later by the *International Cloud Atlas I* (1975), *II* (1987) of the World Meteorological Organization, the *Cloud Studies in Color* by Scorer and Wexler (1967) and the Encyclopedia *Clouds of the World* by Scorer (1972). In this last reference, excellent colored photographs are provided together with a full description of the major genera, species, and varieties of atmospheric clouds. An excellent collection of clouds, photographed from satellites, is found in another book by Scorer (1986).

Both Lamarck and Howard believed the clouds they studied consisted of water bubbles. The bubble idea was originated in 1672 by von Guericke (1602-1686), who called the small cloud particles he produced in a crude expansion chamber 'bullulae' (bubbles). Although he explicitly named the larger particles in his expansion chamber 'guttulae' (drops), the bubble idea, supported by the Jesuit priest Pardies (1701), prevailed for more than a century until Waller (1816-1870) reported in 1846 that the fog particles he studied did not burst on impact, as bubbles would have.

Although this observation was confirmed in 1880 by Dines (1855-1927), it was left to Assmann (1845-1918) to finally end the dispute through the authority of his more comprehensive studies of cloud droplets under the microscope (1884).

The first attempt to measure the size of fog droplets with the aid of a microscope was made by Dines in 1880. Some early measurements of the size of much larger raindrops were made by ingeniously simple and effective means. For example, in 1895 Wiesner (1838-1916) allowed raindrops to fall on filter paper impregnated with water-soluble dye and measured the resulting stains. A little later, Bentley (1904) described an arrangement in which drops fell into a layer of flour and so produced pellets whose sizes could easily be measured and related to the parent drop sizes.

The elegant geometry of solid cloud particles has no doubt attracted attention from the earliest times. Perhaps the first documentation of snow crystals that exhibit a six-fold symmetry was due to an author named Han Ying who made this observation in 1358 BC in China. It was not until centuries later that the same observation also became documented in Europe by means of a woodcut done in 1555 by Olaus Magnus, Archbishop of Uppsala in Sweden. Kepler (1571-1630) was also intrigued by the forms of snow crystals and asked 'Cur autem sexangula?' ('But why are they six-sided?'). Descartes (1596-1650) was perhaps the first to correctly draw the shape of some typical forms of snow crystals (1635). Hooke (1635-1703) first studied the forms of snow crystals under a microscope. Scoresby (1789-1857), in his report on arctic regions (1820), presented the first detailed description of a large number of different snow crystal forms and noticed a dependence of shape on temperature. A dependence of the shape of snow crystals on meteorological conditions was also noted by Martens (1675). Further progress was made when Neuhaus (1855-1915) introduced microphotography as an aid in studying snow crystals. Hellmann (1854-1939) pointed out in 1893 that snow crystals have an internal structure, which he correctly attributed to the presence of capillary air spaces in ice. The most complete collections of snow crystal photomicrographs were gathered by Bentley in the U.S. (published by Humphreys in 1931), and during a life's work by Nakaya (1900–1962) in Japan (published in 1954).

It was also realized early that not all ice particles have a six-fold symmetry. However, before the turn of the 18th century, interest in the large and often quite irregular shaped objects we now call hailstones was apparently restricted to their outward appearance only. Volta (1745-1827) was among the first to investigate their structure, and in 1808 he pointed out that hailstones contain a 'little snowy mass' at their center. In 1814 von Buch (1774-1853) advocated the idea that hailstones originate as snowflakes. This concept was further supported by Waller and Harting (1853), who investigated sectioned hailstones under the microscope. In addition to finding that each hailstone has a center which, from its appearance, was assumed to consist of a few closely-packed snowflakes, they discovered that hailstones also have a shell structure with alternating clear and opaque layers, due to the presence of more or less numerous air bubbles.

All known observations of cloud and precipitation particles were made at ground level until 1783, when Charles (1746-1823) undertook the first instrumented balloon flight into the atmosphere. Although frequent balloon flights were made from that time on, they were confined mostly to studies of the pressure, temperature, and humidity of the atmosphere, while clouds were generally ignored. The first comprehensive study of clouds by manned balloon was conducted by Wigand (1882-1932), who described the in-cloud shape of ice crystals and graupel particles (snow pellets or small hail) in 1903.

Attempts to provide quantitative explanations for the processes which lead to the formation of cloud particles came relatively late, well into the period of detailed observations on individual particles. For example, in 1875 Coulier (1824-1890) carried out the first crude expansion chamber experiment which demonstrated the important role of air-suspended dust particles in the formation of water drops from water vapor. A few years later, Aitken (1839-1919) became the leading advocate of this new concept. He firmly concluded from his experiments with expansion chambers in 1880 that cloud drops form from water vapor only with the help of dust particles which act as nuclei to initiate the new phase. He categorically stated that 'without the dust particles in the atmosphere there will be no haze, no fog, no clouds and therefore probably no rain'. The experiments of Coulier and Aitken also showed that by progressive removal of dust particles by filtration, clouds formed in an expansion chamber became progressively thinner, and that relatively clean air would sustain appreciable vapor supersaturations before water drops appeared. The findings of Coulier and Aitken were put into a more quantitative form by Wilson (1869-1959), who showed in 1897 that moist air purified of all dust particles would sustain a supersaturation of several hundred percent before water drops formed spontaneously. This result, however, was already implicitly contained in the earlier theoretical work of W. Thomson (the later Lord Kelvin, 1824-1907), who showed that the equilibrium vapor pressure over a curved liquid surface may be substantially larger than that over a plane surface of the same liquid (1870).

As soon as experiments established the significant role of dust particles as possible initiators of cloud drops, scientists began to look closer at the nature and origin of these particles. Wilson followed up his early studies with dust-free air and discovered in 1899 that ions promote the condensation process, a result which had been predicted theoretically in 1888 by J.J. Thomson (1856-1940). However, it was soon realized that the supersaturations necessary for water drop formation on such ions were much too large for them to be responsible for the formation of atmospheric clouds. It was again Aitken who noticed in 1881 that due to their different composition, some dust particles seem to be better nuclei than others. He even surmised that 'fine sodic chloride particles' would condense vapor before the vapor was cooled to the saturation point. This observation he attributed to the 'great attraction which salt has for water'. Aitken's observations were further extended by Welander (1897) and Lüdeling (1903), who suggested that Aitken's salt particles are injected into the atmosphere by the world oceans. The great importance of such salt particles to serve as condensation nuclei was also realized by Köhler (1888-1982) who pointed out that the presence of large numbers of hygroscopic particles generally should prevent large supersaturation from occurring in clouds. Also, Köhler was the first to derive a theoretical expression for the variation of vapor pressure over the curved surface of an aqueous solution drop (1921, 1922, 1927). His pioneering studies became the foundation of modern condensation theory.

Although the sigificance of oceans as a souce of condensation nuclei was by now clearly recognized, Wigand's observations (1913, 1930) suggested that the continents, and not the oceans, are the most plentiful source. Wigand's conclusions (1934) were supported by the studies of Landsberg (1906-1985) and Bossolasco (1903-1981).

Lüdeling and Linke (1878-1944) were probably the first to determine the concentration of condensation nuclei in the atmosphere (1903, 1904). However, it was Wigand who, during balloon flights from 1911 to 1913, first carried out detailed studies of condensation nuclei concentrations at different levels in the atmosphere as a function of various meteorological parameters. He discovered that their concentration was related to the temperature structure in the atmosphere, and was significantly different inside and outside clouds. On comparing the concentrations of condensation nuclei and cloud drops, Wigand concluded that there are sufficient numbers of condensation nuclei in the amosphere to account for the number of drops in clouds.

Studies during the same period brought out the fact that dust particles also play an important role in the formation of ice crystals. Thus, those researchers who ascended into clouds with instrumented balloons, found ice crystals at temperatures considerably warmer than the temperatures to which Fahrenheit (1686-1736) had supercooled highly purified water in the laboratory in 1724. Nevertheless, Saussure (1740-1799) pointed out in 1783 that, despite the large number of condensation nuclei, cloud drops generally resist freezing to temperatures much below 0°C. This implied that apparently only a few of the dust particles present in the atmosphere act as ice-forming nuclei. Wegener (1880-1931) suggested that water drops form on water-soluble, hygroscopic nuclei while ice crystals form on a selected group of dust particles which must be water-insoluble. From his observations during a Greenland expedition (1912-1913), he concluded that ice crystals form as a result of the direct deposition of water vapor onto the surface of ice-forming nuclei. He therefore termed this special group of dust particles 'sublimation nuclei'. Wegener's mechanism of ice crystal formation by direct vapor deposition was also advocated by Findeisen (1909-1945). On the other hand, Wigand concluded from his balloon flights that ice crystal formation is often preceded by the formation of supercooled water drops, which subsequently freeze as a result of contact with water-insoluble dust particles. Other arguments against a sublimation mechanism for the formation of ice crystals were brought forward by Krastanow (1908-1977) who, in 1936, theoretically demonstrated that the freezing of supercooled drops is energetically favored over the formation of ice crystals directly from the vapor.

While all these studies provided some answers concerning why and how cloud particles come into being, they did not provide any clues as to how a cloud forms as a whole and why some clouds precipitate and others do not. One of the first precipitation theories was formulated in 1784 by Hutton (1726-1797). He envisioned that the cloud formation requisite to precipitation is brought about by the mixing of two humid air masses of different temperatures. The microphysical details of the apportioning of the liquid phase created by this cooling process were not considered. The meteorologists Dove (1803-1879) and Fitz Roy (1805-1865) evidently were in favor of his theory, since it seemed to predict the observed location of rain at the boundary between 'main currents of air' (this is now interpreted as frontal rain). Therefore, Hutton's precipitation theory persisted for almost a century. When at last given up, it was not for apparent meteorological reasons but for the physical reason that, owing to the large amount of latent heat released during the phase change of water vapor to water, Hutton's process provides far too small an amount of condensed water to explain the observed amounts of rain.

It finally became clear that only cooling by expansion of humid air during its ascent in the atmosphere would provide clouds with sufficient condensed water to account for the observed rain. Thus, Hamberger (1662-1716) noted in 1743 and Franklin (1706-1790) in 1751 that air rises on heating. In turn, Ducarla-Bonifas (1738-1816) and de Saussure (1740-1799) formulated a theory which made use of this concept suggesting that warm moist air which rises will cool as it rises and produce precipitation at a rate which is proportional to the rate of ascent of the moist air. However, it was left to Erasmus Darwin (1731-1802) to clearly formulate in 1788 the connection between expansion, cooling and condensation. The first mathematical formulation of the cooling which is experienced by a volume of expanding air was given by Poisson (1781-1840) in 1823, thus providing the basis for understanding von Guericke's 'cloud chamber' experiments carried out 150 years earlier. Soon afterwards, the idea of cooling by adiabatic expansion, according to which there is no heat exchange between the rising parcel of air and the environment, was applied to the atmosphere by Espy (1785-1860). He deduced in 1835 from experiments and theory that, for a given expansion, dry air is cooled about twice as rapidly as air saturated with water vapor, owing to the heat released by condensing vapor. Also, Péclet (1793-1857) showed in 1843 that the rate of dry adiabatic cooling for a rising air parcel is larger than the cooling usually observed during balloon ascents in the atmosphere.

The first quantitative formulation of the 'saturation adiabatic process', according to which the condensation products are assumed to remain inside the water-saturated air parcel, was worked out by Lord Kelvin in a paper read in 1862 and published in 1865. Meanwhile, in 1864, Reye (1838-1919) independently derived and published formulations for the same process. A mathematical description of the cooling rate of a lifted air parcel from which the condensation products are immediately removed upon formation, a 'pseudoadiabatic process', was formulated in 1888 by von Bezold (1837-1907). In 1884, Hertz (1857-1894) further extended the thermodynamic formulation of a rising moist parcel of air. He suggested that if such a parcel rises far enough, it will pass through four stages: (1) the 'dry stage' in which air is still unsaturated, (2) the 'rain stage' in which saturated water vapor and water are present, (3) the 'hail stage' in which saturated water vapor, water, and ice coexist, and (4) the 'snow stage' in which only water vapor and ice are present.

In 1866, Renou (1815-1902) first pointed out that ice crystals may play an important role in the initiation of rain. Solely on the basis of the rather restricted meteorological conditions he observed, Renou suggested that for development of precipitation, two cloud layers are required: one consisting of supercooled drops and another at a higher altitude which feeds ice crystals into the cloud layer below.

More significant progress in understanding precipitation formation involving ice crystals was achieved by Wegener (1911), who showed through thermodynamic principles that, at temperatures below 0°C, supercooled water drops and ice crystals cannot coexist in equilibrium. Using this result, Bergeron (1891-1977) proposed in 1933 that precipitation is due to the colloidal instability which exists in clouds containing both supercooled drops and ice crystals. Bergeron envisioned that in such clouds the ice crystals invariably grow by vapor diffusion at the expense of the supercooled water drops until either all drops have been consumed or all ice crystals have fallen out of the cloud. Findeisen's cloud observations (1938) produced further evidence in favor of the Wegener-Bergeron precipitation mechanism.

Descartes (1637) had observed that hailstones often have a snowy globule in the middle. In suggesting a mechanism for the formation of hailstones, he therefore speculated that hailstones are the result of numerous snowflakes 'being driven together by wind'. Later, Ducarla Bonifas (1738-1816) proposed with considerable foresight in 1780 that 'columns of air, more strongly heated than the surrounding atmosphere, may violently rise to elevations where the temperature is sufficiently low that the condensation products freeze to become little snowy globules which further grow from the vapor and by collision with supercooled water drops until they are heavy enough to fall back to Earth'. Similarly, von Buch in 1814 and Maille (1802-1882) in 1853 suggested that hailstones originate as snow pellets and grow further by collision with supercooled water drops. Much later, Köhler (1927) applied the notion of collision growth to ice crystals, which he recognized might collect supercooled cloud drops. He also noted, but did not explain, his observation that both drops and crystals have to be of a minimum critical size before such growth may evolve.

The same basic idea of collisional growth, applied this time to cloud drops of different size and hence different fall velocities, was put forth independently in 1715 by Barlow (1639-1719) and by Musschenbroek (1692-1761) in 1739. Musshenbroek also proposed that drops growing by collision will not exceed a size of about 6 mm in diameter, due to the observed instability of drops larger than this size. Reynolds (1842-1912) expanded on the notion of collisional growth and showed by computation in 1877 that water drops above a certain size grow slower by vapor diffusion than by collision with other drops.

A subtle aspect of the collisional growth process was discovered by Lenard (1862-1947), who observed in 1904 that colliding drops do not always coalesce. This he attributed correctly to the difficulty of completely draining all the air from between the colliding drops. He also found (as had been noticed in 1879 by Strutt, later Lord Rayleigh, 1842-1919) that small amounts of electric charge residing on drops could build up attractive electric forces which are sufficiently large to overcome the hydrodynamic resistance to coalescence. In agreement with the expectations of Musschenbroek, Lenard concluded from his experiments that growth by collision-coalescence continues until drops grow to a critical size, after which they become hydrodynamically unstable and break up. He suggested that the fragment drops may then continue to grow in the same manner, producing a 'chain-reaction' effect of overall rapid growth.

Despite Lenard's experimental results, the mechanism of growth by collision was paid little attention for a long time, since the Wegener-Bergeron-Findeisen mechanism dominated the thinking of meteorologists, most of whom studied storm systems at the middle and higher latitudes where the ice phase is quite common. Simpson (1878-1965) attempted to revive the collision mechanism in his presidential address to the Royal Meteorological Society in 1941. On the basis of reports from airplane pilots who flew over India through precipitating clouds with tops thought to be warmer than 0°C, and from some crude calculations made by Findeisen on the rate at which unequal size cloud drops coagulate, Simpson asserted that he found it untenable to assume that precipitation formation should be confined only to clouds which reach subzero temperature levels. However, convincing quantitative support for Simpson's position had to await the late 1940's, when radar observations and military flights finally led to a general consensus that clouds need not reach subzero temperature levels, and consequently need not contain ice crystals for precipitation to occur.

* * *

In striking contrast to the rather slow development of cloud physics prior to 1940, an abrupt and accelerating increase in research and knowledge has occurred since. A confluence of several factors has brought about this dramatic change. For example, a surge of interest in cloud physics was closely tied to the military-related research in meteorology which developed during the war years (1939-1945) and produced a great number of trained workers in meteorology. Also, several new observational techniques involving aircraft, radar, and other instruments became available to scientists at a time when both the necessary funding and support personnel were also relatively abundant. In addition, interest and support was stimulated by the demonstration of Schaefer and Langmuir in 1946 that it is possible to modify at least some clouds and affect their precipitation yield by artificial means. (They seeded supercooled stratus clouds with dry ice, which caused the formation and subsequent rapid growth of ice crystals. This induced colloidal instability led, in about 20 minutes, to a miniature snowfall.) Finally, the fast pace of general technological advances has had a continuing great impact on cloud physics, insuring an accelerated development by making available such important tools as computers, satellites, rockets, and accurately controlled climatic chambers and wind tunnels.

To a large extent, the rapid progress referred to above can be characterized as a fairly direct development of the ideas and discoveries which were made considerably earlier. As we shall see, the period of progress since the beginning of the 1940's has not been characterized by numerous conceptual breakthroughs, but rather by a series of progressively more refined quantitative theoretical and experimental studies of previously identified microphysical processes.

As we shall also see, much remains to be learned in spite of the significant advances of the past four decades. One principal continuing difficulty is that of incorporating, in a physically realistic manner, the microphysical phenomena in the broader context of the highly complex macrophysical environment of natural clouds. This problem was well expressed 35 years ago in the preface to the first edition of Mason's (1957a) treatise on cloud micrphysics.

Although the emphasis here is upon the *micro-physical* processes, it is important to recognize that these are largely controlled by the atmospheric motions which are manifest in clouds. These *macro-physical* features of cloud formation and growth, which might more properly be called a *dynamics*, provide a framework of environmental conditions confining the rates and duration of the micro-physical events. For example, the growth or freezing of cloud droplets is accompanied by the release of great quantities of latent heat, profoundly influencing the motion of cloudy air masses, while the motions which ultimately cause evaporation of the cloud determine its duration, and will set a limit to the size which its particles can attain. Progress in cloud physics has been hindered by a poor appreciation of these interrelations between processes ranging from nucleation phenomena on the molecular scale to the dynamics of extensive cloud systems on the scale of hundreds or thousands of kilometers.

The problem of scale which Mason refers to provides a revealing point of view for appreciating the extent of the difficulties one encounters. Thus, stating the case in a very conservative manner, we are concerned in cloud microphysics with the growth of particles ranging from the characteristic sizes of condensation nuclei ($\geq 10^{-2}\,\mu$m) to precipitation particles ($\leq 10^{4}\,\mu$m for raindrops, $\leq 10^{5}\,\mu$m for hailstone). This means we must follow the evolution of the particle size spectrum, and the attendant microphysical processes of mass transfer, over about seven orders of magnitude in particle size. Similarly, the range of relevant cloud-air motions varies from the characteristic size of turbulent eddies which are small enough to decay directly through viscous dissipation ($\geq 10^{-2}$ cm), since it is these eddies which turn out to define the characteristic shearing rates for turbulent aerosol coagulation processses, to motion on scales at least as large as the cloud itself ($> 10^{5}$ cm). Thus, relevant interactions may occur over at least seven orders of magnitude of eddy sizes. Also, in recent years it has become increasingly clear that a strong coupling may occasionally occur between the particle growth processes, including the development of precipitation, and the growth of the cloud electric field. Since in the atmosphere field strengths range from the fair-weather value ($\leq 10^{2}$ V m^{-1}) to fields of breakdown value (10^{6} V m^{-1}), to understand the formation of highly electrified clouds, we must cope with about four orders of magnitude of electric field variation. At the same time, we also must be concerned with various electrostatic force effects arising from at least an eight order of magnitude range of particle charge, considering the observed presence of 1 to 10^{8} free elementary charges (5×10^{-10} to 5×10^{-2} e.s.u.) on atmospheric particles. If the electrostatic contribution to the large-scale cloud energetics is also considered, a much larger charge magnitude range is involved. Recent studies have shown further that atmospheric clouds and precipitation significantly affect the chemical nature of the atmosphere in that they are able to incorporate aerosol particles as well as certain gaseous atmospheric constituents, which, once dissolved in the drops, allow chemical reactions to alter their chemical nature. Since observations show that the concentration of aerosol particles ranges from a few per cm^{3} in remote background air to a few million per cm^{3} in heavily polluted air over cities, while the concentration of pullutant gases range from a volume fraction of 10^{-10} to one of 10^{-4} in these same locations, we must follow the uptake of atmospheric chemical constituents by clouds and precipitation over about six orders of magnitude of concentration variation.

It is clear, therefore, that a complete in-context understanding of cloud microphysics including dynamic, electrical and chemical effects must await some sort of grand synthesis, an elusive and distant goal even from the point of view of presently available models. We should emphasize that such an approach to the subject is far beyond the scope of this book. Rather, our goal is to provide where possible a reasonably quantitative account of the most relevant, individual microphysical processes. In addition to whatever intrinsic interest and usefulness in other application the separate case studies of this book may hold, we also hope they may help provide a useful basis for an eventual integrated treatment of overall cloud behavior. As we shall see, however, even this restricted approach to the subject necessarily involves a degree of incompleteness, since many microphysical mechanisms are still not understood in quantitative detail. In this sense also cloud microphysics is still a developing subject, and so is characterized to some extent by inadequate knowledge as well as conflicting results and points of view.

CHAPTER 2

MICROSTRUCTURE OF ATMOSPHERIC CLOUDS AND PRECIPITATION

Before discussing the microphysical mechanisms of cloud particle formation, we shall give a brief description of the main microstructural features of clouds. Here we will be concerned primarily with the sizes, number concentrations, and geometry of the particles comprising the visible cloud.

2.1 Microstructure of Clouds and Precipitation Consisting of Water Drops

2.1.1 THE RELATIVE HUMIDITY INSIDE CLOUDS AND FOGS

Although the relative humidity of clouds and fogs usually remains close to 100%, considerable departures from this value have been observed. Thus, reports from different geographical locations (Pick, 1929, 1931; Neiburger and Wurtele, 1949; Mahrous, 1954; Reiquam and Diamond, 1959; Kumai and Francis, 1962a,b) show that the relative humidity of fogs has been found to range from 100% to as low as 81%. Somewhat smaller departures from saturation are usually observed in cloud interiors. Warner (1968a) indirectly deduced values for the relative humidity in small to moderate cumuli from measurements of vertical velocity and drop size. From his results (shown in Figure 2.1), we see that in these clouds the relative humidity rarely surpasses 102% (i.e., a supersaturation of 2%), and is rarely lower than 98%. The median of the observed supersaturations was about 0.1%. Similarly, Braham (in Hoffer, 1960) found, during several airplane traverses through cumulus clouds, that in their outer portions the air generally had relative humidities between 95 and 100%, dipping to as low as 70% near the cloud edges where turbulent mixing was responsible for entraining drier air from outside the clouds. In the more interior cloud portions, the relative humidity ranged from 100% to as high as 107% (shown in Figure 2.2). More recently, Politovich and Cooper (1988) deduced from flights through 147 clouds over Miles City, Montana, that the supersaturation within these clouds ranged between -0.5 and 0.5% with an average of 0%.

Usually, the maximum supersaturation attained for a given updraft in a fog or cloud is inferred from a comparison between the observed number concentration of drops with the observed number concentration of aerosol particles which can form drops at a given supersaturation (Squires, 1952, Warner, 1968a; Hudson, 1980; Meyer *et al.*, 1980; Paluch and Knight, 1984; Austin *et al.*, 1985; Politovich and Cooper, 1988). Recently, however, instruments have become available which are able to measure the relative humidity in clouds more directly. Thus, the relative humidty inside fogs (Figure 2.3) was measured by Gerber (1981) by means of a spe-