THERMODYNAMICS, KINETICS, AND MICROPHYSICS OF CLOUDS

Climate change has provided a new impetus for research on clouds and precipitation. One of the greatest uncertainties in current global climate models is cloud feedback, arising from uncertainties in the parameterization of cloud processes and their impact on the global radiation balance. In the past two decades, substantial progress has been made in the simulation of clouds using cloud resolving models. However, most of the parameterizations employed in these models have been empirically based. New theoretical descriptions of cloud processes are now being incorporated into cloud models, using spectral microphysics based on the kinetic equations for the drop and crystal size spectra along with the supersaturation equation, and newer parameterizations of drop activation and ice nucleation based on the further development of the classical nucleation theory. From these models, cloud microphysics parameterizations are being developed for use in global weather and climate models.

Thermodynamics, Kinetics, and Microphysics of Clouds reflects this shift to an increasingly theoretical basis for the simulation and parameterization of cloud processes. The book presents a unified theoretical foundation that provides the basis for incorporating cloud microphysical processes in cloud and climate models in a manner that represents interactions and feedback processes over the relevant range of environmental and parametric conditions. In particular, this book provides:

- the closed system of equations of spectral cloud microphysics that includes kinetic equations for the drop and crystal size spectra for regular and stochastic condensation/deposition and coagulation/accretion along with the supersaturation equations;
- the latest theories and theoretical parameterizations of aerosol hygroscopic growth, drop activation, and ice homogeneous and heterogeneous nucleation, derived from the general principles of thermodynamics and kinetics and suitable for cloud and climate models;
- a theoretical basis for understanding the processes of cloud particle formation, evolution, and precipitation, based on numerical cloud simulations and analytical solutions to the kinetic equations and supersaturation equation;
- a platform for advanced parameterizations of clouds in weather prediction and climate models using these solutions; and
- the scientific foundation for weather and climate modification by cloud seeding.

This book will be invaluable for researchers and advanced students engaged in cloud and aerosol physics, and air pollution and climate research.

Vitaly I. Khvorostyanov is Professor of Physics of the Atmosphere and Hydrosphere, Central Aerological Observatory (CAO), Russian Federation. His research interests are in cloud physics, cloud numerical modeling, atmospheric radiation, and cloud-aerosol and cloud-radiation interactions, with applications for climate studies and weather modification. He has served as Head of the Laboratory of Numerical Modeling of Cloud Seeding at CAO, Coordinator of the Cloud Modeling Programs on Weather Modification by Cloud Seeding in the USSR and Russia, Member of the International GEWEX Radiation Panel of the World Climate Research Program, and Member of the International Working Group on Cloud-Aerosol Interactions. Dr. Khvorostyanov has worked as a visiting scientist and Research Professor in the United States, United Kingdom, France, Germany, and Israel. He has co-authored nearly 200 journal articles and four books: *Numerical Simulation of Clouds* (1984), *Clouds and Climate* (1986), *Energy-Active Zones: Conceptual Foundations* (1989), and *Cirrus* (2002). Dr. Khvorostyanov is a member of the American Geophysical Union and the American Meteorological Society.

Judith A. Curry is Professor and Chair of the School of Earth and Atmospheric Sciences at the Georgia Institute of Technology. She previously held faculty positions at the University of Colorado, Penn State University, and Purdue University. Dr. Curry's research interests span a variety of topics in the atmospheric and climate sciences. Current interests include cloud microphysics, air and sea interactions, and climate feedback processes associated with clouds and sea ice. Dr. Curry is co-author of *Thermodynamics of Atmospheres and Oceans* (1999) and editor of the *Encyclopedia of Atmospheric Sciences* (2003). She has published more than 190 refereed journal articles. Dr. Curry is a Fellow of the American Meteorological Society, the American Association for the Advancement of Science, and the American Geophysical Union. In 1992, she received the Henry Houghton Award from the American Meteorological Society.

The Cloud
Percy Bysshe Shelley (1820)

I bring fresh showers for the thirsting flowers,
From the seas and the streams;
I bear light shade for the leaves when laid
In their noonday dreams.
From my wings are shaken the dews that waken
The sweet buds every one,
When rocked to rest on their mother's breast,
As she dances about the sun.
I wield the flail of the lashing hail,
And whiten the green plains under,
And then again I dissolve it in rain,
And laugh as I pass in thunder.

I am the daughter of Earth and Water,
And the nursling of the Sky;
I pass through the pores of the oceans and shores;
I change, but I cannot die.
For after the rain, when with never a stain
The pavilion of Heaven is bare
And the winds and sunbeams with their convex gleams
Build up the blue dome of air,
I silently laugh at my own cenotaph
And out of the caverns of rain,
Like a child from the womb, like a ghost from the tomb,
I arise and unbuild it again.

(Poetical Works of Shelley (Cambridge Editions),
by Percy Bysshe Shelley (Author), Newell F. Ford (Introduction).
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Preface

Cloud microphysics is a branch of cloud physics that studies initiation, growth, and dissipation of cloud and precipitation particles. Cloud microphysics is governed by the thermodynamic and kinetic processes in clouds. The field of cloud microphysics has been intensively developed since the 1940s when the first successful experiments on cloud seeding were performed. The field has received additional impetus in recent years from the challenges associated with forecasting precipitation and understanding aerosol-cloud interactions in the context of climate change and feedback processes. Several books on cloud microphysics are available, including Mason (1957), Fletcher (1962, 1970a), Dufour and Defay (1963), Sedunov (1974), Voloshchuk and Sedunov (1975), Voloshchuk (1984), Matveev (1984), Young (1993), Pruppacher and Klett (1997), and Cotton et al. (2011).

Thermodynamics, Kinetics, and Microphysics of Clouds extends the subject of cloud microphysics beyond these previous treatments. The goals and contents of this book are formulated to:

- Present in compact form the major thermodynamic relations and kinetic equations required for theoretical consideration of cloud microphysics;
- Review the currently known states of water in liquid, crystalline, and amorphous forms, and the conceptual modern theories of water and equations of state for water in various states;
- Formulate a closed system of equations that describe the kinetics of cloud microphysical processes and is suitable both for analytical studies and for inclusion in numerical models;
- Derive from theory generalized analytical parameterizations for aerosol deliquescence, hygroscopic growth, efflorescence, and drop activation and ice nucleation in various modes;
- Demonstrate that these theoretical parameterizations generalize and unify previous parameterizations and include them as particular cases; express previous empirical parameters via atmospheric and aerosol parameters and theoretical quantities;
- Derive the kinetic equations of stochastic condensation and coagulation and obtain their analytical solutions that reproduce the observed drop and crystal size spectra; express parameters of empirical distributions from theory; and
- Outline a path for future generalizations of the kinetic equations of cloud microphysics based on the Chapman–Kolmogorov and Fokker–Planck equations.

Using the general principles of thermodynamics and kinetics, a closed system of equations is formulated that includes kinetic equations for the drop and crystal size spectra along with the supersaturation equations. Using these equations and further developing classical nucleation theory, theories are developed of aerosol hygroscopic growth, drop activation, and ice homogeneous and heterogeneous nucleation. Analytical expressions are obtained for the particle concentration, critical radii and energies of nucleation, nucleation rates that are expressed as functions of temperature, saturation ratio, pressure, and aerosol concentration simultaneously and in factorized form. It is shown that the new theoretical expressions generalize previous empirical parameterizations, can reduce to them in some particular cases, and their empirical parameters are expressed via the aerosol parameters and physical constants. The validity of these new theoretical expressions is verified in comparison with experimental data, previous empirical and semi-empirical parameterizations, and parcel model simulations. A similar theory is developed for the aerosol deliquescence and efflorescence. This allows for the first time calculation from the theory of a unified phase diagram for solutions that are in agreement with experimental phase diagrams.

Various analytical solutions to the kinetic equations and supersaturation equations are obtained for adiabatic and non-adiabatic processes. These solutions are suitable both for analytical studies of condensation and for inclusion in the numerical models. This system of equations, including kinetic equations for drops and crystals and integral supersaturation equations, is generalized for the turbulent atmosphere and multidimensional models. A fast algorithm for a numerical solution based on the splitting method is described. Spectral bin microphysical method was applied for many years in various 1D, 2D, and 3D models for various cloud types, and its applicability in the models of various scales and dimensions is discussed.

The kinetic equations of stochastic condensation in a turbulent atmosphere are derived and generalized taking into account the coagulation and accretion processes. Various analytical solutions to these stochastic equations are obtained, whose functional forms are similar to the gamma distributions and to exponential and inverse power laws that have been observed in clouds and precipitation. The solution parameters are expressed via the atmospheric characteristics and physical constants, and the solutions are verified through comparison with experimentally observed size spectra. These solutions provide explanations of various empirical parameterizations and a platform for their refinement.

In addition to advancing our basic understanding of cloud microphysical processes, the theoretical approach employed in this book supports the explanation and interpretation of laboratory and field measurements in the context of instrument capabilities and limitations and motivates the design of future laboratory and field experiments. In the context of models that include cloud processes, ranging from small-scale models of clouds and atmospheric chemistry to global weather and climate models, the unified theoretical foundations presented here provide the basis for incorporating cloud microphysical processes in these models in a manner that represents the process interactions and feedback processes over the relevant range of environmental and parametric conditions. Further, the analytical solutions presented here provide the basis for computationally efficient parameterizations that include the relevant parametric dependencies. The methods of cloud simulation using spectral bin microphysics described here are especially suitable for modeling of weather modification by cloud seeding since these methods are almost always based on modification of cloud microstructure and phase state. These methods are also convenient for studies of inadvertent cloud modification by anthropogenic and natural pollutions and for studies of cloud-radiation interactions.

This book incorporates the heritage of Russian cloud physics that introduced and developed the kinetic equations for drop and crystal diffusion growth, the fast numerical algorithms for their solutions, and the stochastic approach to cloud microphysical processes. This Russian heritage is combined with the best knowledge of cloud microphysics acquired and described in the Western literature over the past several decades. A large amount of the material presented in this book is based on original work conducted jointly by the authors over almost two decades. Some of this research has been published previously in journal articles, but a large portion of this material is being published here in this book for the first time, notably the parameterization of heterogeneous ice nucleation and the theory of aerosol deliquescence and efflorescence.

Integration of Russian and Western perspectives on cloud physics was facilitated by the 1972 bilateral treaty between the U.S. and USSR on Agreement and Cooperation in the Field of Environmental Protection, specifically under Working Group VIII – The Influence of Environmental Change on Climate. Its regular meetings and exchanges of delegations and information promoted international collaboration, provided the foundation for long-term cooperation, and outlined proposals for joint research. With the advent of the World Climate Research Programme (WCRP) in 1980, both Khvorostyanov and Curry subsequently became members of the WCRP Working Group on Radiative Fluxes, which later became the Radiation Panel of the Global Water and Energy Exchange Experiment (GEWEX). The GEWEX Radiation Panel had regular annual meetings (where the authors participated and met), which initiated the collaboration that has lasted for almost two decades, resulted in more than 30 joint publications, and culminated in this book.

This book bridges Russian and Western perspectives of cloud physics. Khvorostyanov's involvement in the evolution of the Russian school of cloud physics includes development of cloud models with spectral bin microphysics and applications to cloud seeding and cloud-radiation interactions. Curry's early cloud microphysics research focused on aircraft observations of cloud microphysics and the development of parameterizations for cloud and climate models. Over the past 18 years, Khvorostyanov and Curry have collaborated on a range of cloud microphysical topics of relevance to understanding and parameterizing cloud processes for cloud and climate models, that integrate the Russian perspectives on cloud microphysics into the broader community, and that combine Eastern and Western approaches to cloud microphysics. In addition to summarizing and integrating these perspectives and the broad body of recent research in cloud microphysics, throughout the book a number of new results are included, as well as extensions and generalizations of existing ones.

This monograph is intended to provide a source of information for scientists engaged in teaching and research in cloud physics and dynamics, aerosol physics, air pollution, and weather modification. The book can be used as a textbook to provide graduate-level students with the theoretical foundations of cloud microphysics. Researchers and students should have a basic background in physics and thermodynamics and mathematical physics before using this book. Beyond this basic background, the authors have made every effort to make the book as self-inclusive as possible. Formal derivations and analytical solutions are emphasized, with every effort made to make the mathematical steps easy to follow, including additional details in the appendices. A comprehensive bibliography is provided that references seminal material in the primary literature and previous textbooks and monographs.

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