### TABLE OF CONTENTS

PREFACE TO THE FIRS	Γ EDITION <b>Χν</b>
---------------------	---------------------

PREFACE TO	THE SECOND	EDITION	i
PREFACE TO	THE SECOND	EDITION	

# CHAPTER 1 HISTORICAL REVIEW

1

10

## CHAPTER 2

# MICROSTRUCTURE OF ATMOSPHERIC CLOUDS AND PRECIPITATION

<b>2.1</b>	Micros	structure of Clouds and Precipitation Consisting of Water Drops	10
	2.1.1	The Relative Humidity inside Clouds and Fogs	10
	2.1.2	Microstructure of Fogs	12
	2.1.3	Microstructure of Clouds	15
	2.1.4	Formulations for the Drop Size Distributions in Clouds and Fogs	24
	2.1.5	The Mean Distance between Drops in Clouds and Fogs	27
	2.1.6	Microstructure of Rain	30
2.2	Micros	structure of Clouds and Precipitation Consisting of Ice Particles	38
	2.2.1	Shape, Dimensions, Bulk Density and Number	
		Concentration of Snow Crystals	40
	2.2.2	Shape, Dimensions, Bulk Density, and Number	
		Concentration of Snowflakes, Graupel, and Hailstones	58

### **CHAPTER 3**

### THE STRUCTURE OF WATER SUBSTANCE 74

3.1	Struct	ure of an Isolated Water Molecule	'4
		ure of Water Vapor	
		ure of Ice	
3.4	Struct	ure of Water and Aqueous Solutions8	36
	3.4.1	Structure of Water	36
	3.4.2	Structure of Aqueous Solutions	8

### EQUILIBRIUM BETWEEN WATER VAPOR, WATER, AQUEOUS SOLUTIONS, AND ICE IN BULK 100

4.1	Useful Thermodynamic Relations 100
4.2	General Conditions for Equilibrium 102
4.3	Phase Rule for Bulk Phases
4.4	Ideal versus Real Behavior of Dry Air, Water Vapor, and Moist Air105
4.5	Chemical Potential of Water Vapor in Humid Air, and of Water in
	Aqueous Solutions 107
4.6	Equilibrium Between an Aqueous Salt Solution and Water Vapor 109
4.7	Latent Heat of Phase Change and its Temperature Variation 115
4.8	Clausius-Clapeyron Equation116
4.9	Equilibrium Between an Aqueous Salt Solution and Ice

### **CHAPTER 5**

#### SURFACE PROPERTIES OF WATER SUBSTANCE 126

5.1	Surface Tension		
5.2	Equilibrium Conditions		
5.3	Phase Rule for Systems with Curved Interfaces		
5.4	Water-Vapor Interface		
	5.4.1 Effect of Temperature on the Surface Tension of		
	5.4.2 Surface Tension of Aqueous Salt Solutions		
	5.4.3 Radius Dependence of Surface Tension		
5.5	Angle of Contact		
5.6	Adsorption of Water Vapor on Solid Surfaces		
5.7	Ice-Vapor Interface		
	5.7.1 Surface Energy of Ice		
	5.7.2 Wulff's Theorem		
	5.7.3 Structure of Real Ice Surfaces		
5.8	Adsorption of Reactive Gases on Ice Surfaces		
5.9	Ice-Water Interface		
5.10	Ice Aqueous Solution Interface		
5.11	Condensation, Deposition, and Thermal Accommodation Coefficients 163		

### **CHAPTER 6**

# EQUILIBRIUM BEHAVIOR OF CLOUD DROPS AND ICE PARTICLES

167

6.1	General Equilibrium Relation for Two Phases Separated by a	
_	Curved Interface	. 167
6.2	Effect of Curvature on Latent Heat of Phase Change	. 168

6.3	Generalized Clausius-Clapeyron Equation	169
6.4	Equilibrium Between a Pure Water Drop and Pure Water Vapor or	
	Humid Air	170
6.5	Equilibrium Between an Aqueous Solution Drop and Humid Air	172
6.6	Equilibrium Between Humid Air and an Aqueous Solution Drop	
	Containing a Solid Insoluble Substance	175
6.7	Equilibrium Conditions for Ice Particles	178
6.8	Experimental Verification	184

### HOMOGENEOUS NUCLEATION 191

7.1	Homo	geneous Nucleation of Water Drops and Ice Crystals from	
	Water	Vapor	192
	7.1.1	Equilibrium Population of Embryos and Energy of Embryo	
		Formation	192
		7.1.1.1 Formal Statistical Mechanics Description	192
		7.1.1.2 Molecular Model Method	194
		7.1.1.3 The Classical Description	194
	7.1.2	The Nucleation Rate J	199
	7.1.3	Experimental Verification	204
7.2	Homo	geneous Nucleation of Ice in Supercooled Water	205
	7.2.1	The Nucleation Rate J	205
	7.2.2	The Energy of Germ Formation	207
		7.2.2.1 Classical Model	207
		7.2.2.2 The Molecular Model	207
	7.2.3	The Molar Activation Energy $\Delta g^{\ddagger}$	209

## **CHAPTER 8**

# THE ATMOSPHERIC AEROSOL AND TRACE GASES 216

8.1	Gaseo	us Constituents of the Atmosphere	216
8.2	Atmos	pheric Aerosol Particles (AP)2	25
	8.2.1	Formation of Aerosol Particles by Gas to Particle	
		Conversion (GPC)	26
	8.2.2	Formation of Aerosol Particles by Drop Particle Conversion	
		(DPC)	233
	8.2.3	Formation of Aerosol Particles by Bulk to Particle	
		Conversion (BPC)	240
		8.2.3.1 BPC at the Solid Earth Surface	240
		8.2.3.2 BPC at the Surface of Oceans	243
	8.2.4	AP from Extraterrestrial Sources	247
	8.2.5	Rate of Emission of Particulate Matter into the Atmosphere 2	248
	8.2.6	Residence Time of AP	248

8.2.7	Water-se	oluble Fraction of AP	251
8.2.8		ass and Number Concentration of AP	
	8.2.8.1	Number Concentration (except Polar Aerosols).	
	8.2.8.2		
	8.2.8.3	Total Mass and Number Concentration of	
		Particles in Polar, Tropospheric Aerosols	
8.2.9	Size Dis	tribution of AP	
8.2.10		Variation of the Number and Mass Concentration	

# HETEROGENEOUS NUCLEATION 287

9.1	Cloud	Condensation Nuclei (CCN)	287
	9.1.1	Number Concentration and Chemical Composition of CCN.	287
	9.1.2	Mode of Action of Water-Soluble and Mixed CCN	296
	9.1.3	Nucleation of Drops on Water-Insoluble CCN	297
		9.1.3.1 Nucleation on a Planar Substrate	298
		9.1.3.2 Nucleation on a Curved Substrate	302
	9.1.4	Experimental Verification of Heterogeneous Water Drop	
		Nucleation	306
9.2	Ice For	rming Nuclei (IN)	309
	9.2.1	Number Concentration of IN	309
	9.2.2	Sources and Chemical Composition of IN	317
	9.2.3	The Main Requirements for IN	326
		9.2.3.1 Insolubility Requirement	326
		9.2.3.2 Size Requirement	326
		9.2.3.3 Chemical Bond Requirement	328
		9.2.3.4 Crystallographic Requirement	329
		9.2.3.5 Active-Site Requirement	330
	9.2.4	Theory of Heterogeneous Ice Nucleation	341
		9.2.4.1 The Classical Model	341
		9.2.4.2 Extensions of the Classical Model	344
		9.2.4.3 The Semi-Empirical Statistical Mechanics Model	345
	9.2.5	Heterogeneous Freezing of Supercooled Water Drops	
	9.2.6	Discrepancy Between the Concentrations of IN and the	
		Concentration of Ice Particles	355

# CHAPTER 10

# HYDRODYNAMICS OF SINGLE CLOUD<br/>AND PRECIPITATION PARTICLES361

10.1	Basic Governing Equations	
10.2	Flow Past a Rigid Sphere	
	10.2.1 Classification of Flows According to Reynolds Number	364

	10.2.2	Steady, Axisymmetric Flow	366
	10.2.2	10.2.2.1 The Stream Function	
		10.2.2.2 The Drag Problem	
		10.2.2.3 Analytical Solutions	
		10.2.2.4 Numerical Approach to the Navier-Stokes Equation	ı <b>37</b> 8
		10.2.2.5 Comparison of Analytical and Numerical	
		Solutions of the Navier-Stokes Equation with	
		Experimental Results	379
	10.2.3	The Fall Behavior of Rigid Spheres	384
	10.2.4	Non-Steady Three-Dimensional Flow	384
10.3	Hydrod	lynamic Behavior of Water Drops in Air	385
	10.3.1	Internal Circulation in Drops	386
	10.3.2	Drop Shape	393
	10.3.3	Drop Oscillation	400
	10.3.4	Fall Behavior of Drops	409
	10.3.5	Drop Instability and Breakup	410
	10.3.6	Terminal Velocity of Water Drops in Air	
10.4	Hydrod	lynamic Behavior of Disks, Oblate Spheroids, and Cylinders	
	10.4.1	•	
	10.4.2	Circular Cylinders	
10.5	Hydrod	lynamic Behavior of Snow Crystals, Snow Flakes, Graupel	
	-	ilstones	433
	10.5.1	Flow Field and Drag	433
	10.5.2	Fall Velocity	
	10.5.3	Fall Pattern	

# MECHANICS OF THE ATMOSPHERIC AEROSOL 447

11.1	Brownian Motion of Aerosol Particles	447
11.2	Particle Diffusion	449
11.3	Mobility and Drift Velocity	450
11.4	Sedimentation and the Vertical Distribution of Aerosol Particles	451
11.5	Brownian Coagulation of Aerosol Particles	454
11.6	Laminar Shear, Turbulence, and Gravitational Coagulation	463
	11.6.1 Coagulation in Laminar Shear Flow	463
	11.6.2 Coagulation in Turbulent Flow	465
	11.6.2.1 Turbulent Shear Coagulation	467
	11.6.2.2 Turbulent Inertial Coagulation	468
	11.6.3 Gravitational Coagulation	469
11.7	Explanation for the Observed Size Distributions of the Atmospheric	
	Aerosol	472
	11.7.1 Quasi-Stationary Distributions (QSD)	472
	11.7.2 Self-Preserving Distributions (SPD)	474
	11.7.3 Quasi-Stationary Self-Preserving Distributions	480

11.7.4	Statistical Distributions	. 481
11.7.5	Power Law Solutions for a Source-Enhanced Aerosol	. 482

### COOLING OF MOIST AIR

485

12.1	Water in the Atmosphere	. 485
12.2	Isobaric Cooling	. 488
12.3	Adiabatic Cooling of Unsaturated Air	. 488
12.4	The Thermodynamic Wetbulb Temperature	. <b>49</b> 0
19.5	Lifting to Saturation and Beyond	. 490
12.6	Adiabatic Cooling of Saturated Air	. 492
12.0	Cooling with Entrainment	.492
12.8	The Concept of Entrainment	. 493
12.9	The Air Parcel Model for a Convective Cloud	. 497

### CHAPTER 13

# DIFFUSION GROWTH AND EVAPORATION OF WATER DROPS AND SNOW CRYSTALS 502

13.1	Laws fo	r Diffusion of Water Vapor and Heat	502
-	13.1.1	Diffusion of Water Vapor	502
	13.1.2	Diffusion of Heat	507
13.2	Growth	of Aqueous Solution Drops by Diffusion of Water Vapor	509
	13.2.1	Growth of an Individual Stationary Drop	509
		Diffusional Growth of a Population of Solution Drops of	
		Neglible Fall Velocity	512
		13.2.2.1 Condensation Growth in Cumuliform Clouds	512
		13.2.2.2 Condensation Growth in Stratiform Clouds and Fog	
	13.2.3		
		Subsaturated Air	537
13.3	Growth	of Snow Crystals by Diffusion of Water Vapor	
	13.3.1	Growth of a Stationary Snow Crystal	546
	13.3.2	Growth of a Ventilated Snow Crystal	550
		Growth Rate of Snow Crystal Faces - Snow Crystal Habit	
		Change	561

### CHAPTER 14

# CLOUD PARTICLE INTERACTIONS 568

14.1	The Basic Model for Drop Collisions	568
14.2	Definition of Collision Efficiency	569
14.3	The Superposition Method	571

14.4	The Br	oundary Value Problem Approach	574
11.1	14.4.1	The Quasi-Stationary Assumption	
		• • •	
	14.4.2	Two Spheres in Steady Stokes Flow	
	14.4.3	The Slip-Flow Correction in Stokes Flow	577
	14.4.4	Two Spheres in Modified Oseen Flow	
14.5	Collisio	on of Water Drops with Water Drops	581
	14.5.1	The Case of Calm Air	
	14.5.2	The Case of Turbulent Air	
	14.5.3	Experimental Verification	591
	14.5.4	Coalescence of Water Drops in Air	594
		14.5.4.1 The Rebound Problem	595
		14.5.4.2 Disruption Following Collision	598
14.6	Collisio	on of Snow Crystals with Water Drops	599
	14.6.1	Collision of Large Snow Crystals with Small Drops	599
	14.6.2	Collision of Large Drops with Small Snow Crystals	606
14.7	Collisic	on of Snow Crystals with Snow Crystals	
14.8	Orienta	ation Model for Particles in Turbulence	610
	14.8.1	Turbulence Model	
	14.8.2	Orientation of Spheroids in Turbulent Air	611
	14.8.3	Generalized Orientation Distribution	

## GROWTH OF CLOUD DROPS BY COLLISION, COALESCENCE AND BREAKUP

617

15.1	Contin	uous Model for Collection Growth	
15.2	Polyno	mial Approximations to the Gravitational Collection Kernel 620	
15.3	Stocha	stic Model for Collisional Growth	
	15.3.1	Completeness of the SCE	
		15.3.1.1 Three Models for Collection Growth	
		15.3.1.2 Correlations in a Stochastic Coalescence Process 628	
		15.3.1.3 Monte Carlo Study of Stochastic Correlation	
	15.3.2	Exact Solutions to the SCE	
	15.3.3	Numerical and Approximation Techniques for the SCE	
		15.3.3.1 The Method of Berry (1967) and Reinhardt (1972)637	
		15.3.3.2 The Method of Moments	
15.4	Stocha	stic Model for Drop Breakup645	
15.5		ochastic Drop Growth in Combination with Stochastic Drop Breakup 650	

### **CHAPTER 16**

## GROWTH OF ICE PARTICLES BY ACCRETION AND ICE PARTICLE MELTING 659

	16.1.1	Growth Mode and Structure of Rimed Ice Particles,	
		Graupel, and Hailstones	. 659
	16.1.2	Structure and Growth Mode of Ice in Supercooled Water	. 663
		Growth Rate of Ice in Supercooled Water	
	16.1.4	Freezing Time of Water Drops	. 674
		Growth Rate of Graupel and Hailstones	
		Snow Crystal Multiplication by Riming	
16.2	Growth	of Snow Crystals by Collision with other Snow Crystals	. 689
16.3	Melting	of Ice Particles	. 691
	16.3.1	Melting of Graupel and Hailstones	. <b>692</b>
	16.3.2	Melting of Snow Flakes	. 697

# CLOUD CHEMISTRY

700

17.1	Concen	trations of Water Soluble Compounds in Bulk Cloud and		
	Rain Water, and in Bulk Water of Melted Snow		. 701	
17.2	Concen	tration of Water Insoluble Particles in Bulk Cloud and Rain		
	Water a	and Bulk Water of Melted Snow	. 708	
17.3		atration of Water Soluble Compounds in Individual Cloud		
	and Ra	undrops	. 711	
17.4		ging of Aerosol Particles by Cloud Drops, Raindrops and Ice		
	Particle	es	. 715	
	17.4.1	Nucleation Scavenging	. 716	
	17.4.2	Impaction Scavenging	. 720	
		17.4.2.1 Scavenging by Convective Brownian Diffusion	720	
		17.4.2.2 Scavening by Thermophoresis and Diffusiophoresis	. 724	
		17.4.2.3 Scavenging by Gravitational or Inertial Impaction	730	
		17.4.2.4 Scavenging by Turbulence	. 732	
		17.4.2.5 Combined Force Effects: the Trajectory and Flux		
		Models	732	
17.5	Scaven	iging of Gases by Cloud Drops, Raindrops and Ice Particles	744	
	17.5.1	Scavenging of Gases by Water Drops	. 749	
		17.5.1.1 Solution and Dissociation Equilibria	749	
		17.5.1.2 Diffusion Models for Gases	. 757	
	17.5.2	17.5.2 Asymmetry between Absorption and Desorption of Gases7		
	17.5.3	Deviations from Equilibrium	777	
	17.5.4	Scavenging of Gases by Ice Particles	783	
17.6	The Se	cavenging Parameters	784	
17.7	Wet D	Deposition	787	

# **CHAPTER 18**

CLOUD	ELECTRICITY	792
-------	-------------	-----

18.1	Electrical State of the Cloudless	Atmosphere	192
------	-----------------------------------	------------	-----

18.2	Electric	ctrical State of the Atmospheric Aerosol		
18.3	Electric	cal Conductivity in Clouds	798	
	18.3.1	Diffusion and Conduction of Ions to Cloud Drops	798	
	18.3.2	Conductivity in Weakly Electrified Clouds	799	
	18.3.3	Conductivity in Strongly Electrified Clouds		
18.4	Charge	Distribution in Clouds		
	18.4.1	Weakly Electrified Clouds		
	18.4.2	Strongly Electrified Clouds	806	
18.5	Cloud (	Charging Mechanisms	811	
	18.5.1	Requirements for a Cloud Charging Mechanism		
	18.5.2	The Major Cloud Charging Mechanisms		
		18.5.2.1 Charging by Diffusion of Ions	812	
		18.5.2.2 Convection Charging	812	
		18.5.2.3 Inductive Charging Mechanisms	816	
		18.5.2.4 Non-Inductive Charging Mechanisms Involving		
		the Collision between Particles	821	
		18.5.2.5 Non-Inductive Charging Mechanisms Involving		
		the Breakup of Precipitation Particles	824	
		18.5.2.6 Growth of the Electric Field	826	
18.6	Effect o	f Electric Fields and Charges on Microphysical Processes	827	
	18.6.1	Drop and Ice Crystal Nucleation	827	
	18.6.2	Diffusional Growth of Ice Crystals	828	
	18.6.3	Drop Deformation, Disruption and Corona Production	829	
	18.6.4	Drop Terminal Velocities	835	
	18.6.5	Collisional Growth Rate of Cloud Particles	836	
	18.6.6	Scavenging of Aerosol Particles	846	
APPEN	NDICES		853	
REFEF	RENCES .	•••••••••••••••••••••••••••••••••••••••	874	
LIST OF PRINCIPAL SYMBOLS			935	
TABLE	OF PHY	YSICAL CONSTANTS	944	
INDEX			945	