Modeling of Atmospheric Chemistry

Mathematical modeling of atmospheric composition is a formidable scientific and computational challenge. This comprehensive presentation of the modeling methods used in atmospheric chemistry focuses on both theory and practice, from the fundamental principles behind models, through to their applications in interpreting observations. An encyclopedic coverage of methods used in atmospheric modeling, including their advantages and disadvantages, makes this a one-stop resource with a large scope. Particular emphasis is given to the mathematical formulation of chemical, radiative, and aerosol processes; advection and turbulent transport; emission and deposition processes; as well as major chapters on model evaluation and inverse modeling. The modeling of atmospheric chemistry is an intrinsically interdisciplinary endeavor, bringing together meteorology, radiative transfer, physical chemistry, and biogeochemistry. This book is therefore of value to a broad readership. Introductory chapters and a review of the relevant mathematics make the book instantly accessible to graduate students and researchers in the atmospheric sciences.

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"This exceptional volume by two pioneers in the field covers every essential aspect of atmospheric modeling."

- John Seinfeld, *California Institute of Technology*

"An impressive and comprehensive description of the theoretical underpinning and practical application of atmospheric chemistry modeling. Soon to be a classic reference for graduate students and researchers in the field."

- Colette L. Heald, *Massachusetts Institute of Technology*

"Brasseur and Jacob, both world leaders in modeling atmospheric chemistry, have written a thoroughly engaging textbook. The breadth and depth of the material covered in the book is impressive, but a major strength of the book is the ability of the authors to present often complex information in an accessible way. I have no doubt that this book will help educate future generations of scientists and be a reference point for researchers worldwide. It will certainly become a well-thumbed volume on my bookshelf.

- Paul Palmer, *University of Edinburgh*

"This excellent book provides a comprehensive introduction and reference to modeling of atmospheric chemistry from two of the pioneering authorities in the field. From the historical motivations through to modern-day approaches, the atmospheric physical, chemical and radiative components of the model framework are described. What makes this book particularly relevant and timely is the discussion of the methods for integrating observations and models that are at the forefront of current scientific advancement."

- David P. Edwards, *National Center for Atmospheric Research*

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Preface

Modern science dealing with complex dynamical systems increasingly makes use of mathematical models to formalize the description of interactive processes and predict responses to perturbations. Models have become fundamental tools in many disciplines of natural sciences, engineering, and social sciences. They describe the essential aspects of a system using mathematical concepts and languages and they can in this manner provide powerful approximations of reality. They are used to analyze observations, understand relationships, test hypotheses, and project future evolution. Disagreements between models and observations often lead to important advances in theoretical understanding. Models also play a critical role in the development of policy options and in decision-making.

In atmospheric science, mathematical models have long been central tools for weather prediction and climate research. They are now also used extensively to describe the chemistry of the atmosphere. The corresponding model equations describe the factors controlling atmospheric concentrations of chemical species as a function of emissions, transport, chemistry, and deposition. Chemical species are often coupled through intricate mechanisms, and the corresponding differential equations are then also coupled. Simulation of aerosol particles needs to account in addition for microphysical processes governing particle size and composition, as well as interactions with the hydrological cycle through cloud formation. The difficulty of modeling atmospheric composition is compounded by the need to resolve a continuum of temporal and spatial scales stretching over many orders of magnitude from microseconds to many years, from local to global, and involving coupling of transport and chemistry on all scales.

Mathematical modeling of atmospheric chemistry is thus a formidable scientific and computational challenge. It integrates elements of meteorology, radiative transfer, physical chemistry, and biogeochemistry. Solving the large systems of coupled nonlinear partial differential equations that characterize the atmospheric evolution of chemical species requires advanced numerical algorithms and pushes the limits of supercomputing resources.

The purpose of this book is to provide insight into the methods used in models of atmospheric chemistry. The book is designed for graduate students and professionals in atmospheric chemistry, but also more broadly for researchers interested in atmospheric models, numerical methods, and optimization theory.

The book is divided into three parts. The first part presents background material. Chapter 1 introduces the reader to the concept of model and provides a historical perspective on the development of atmospheric and climate models, leading to the development of atmospheric chemistry models. It reviews the different types of atmospheric chemistry models and highlights their role as components of observing systems.

Fundamentals of atmospheric dynamics and chemistry are presented in Chapters 2 and 3. Chapter 2 describes the vertical structure of the atmosphere, defines key parameters that characterize the dry and the wet atmosphere, and introduces the concept of static stability and geostrophic balance. It goes on to describe the general circulation of the atmosphere. Chapter 3 provides a summary survey of the chemical processes relevant to the atmosphere as well as the microphysical processes controlling the evolution of aerosol particles. Chapter 4 presents the fundamental mathematical equations on which atmospheric models are based and gives an introduction to the numerical methods used to solve these equations.

The second part of this book focuses on the formulation of model processes and reviews the numerical algorithms used to solve the model equations. Chapter 5 covers the formulation of radiative transfer, chemical kinetics, and aerosol microphysics. Chapter 6 reviews numerical methods to solve the stiff systems of nonlinear ordinary differential equations that describe atmospheric chemistry mechanisms. Chapter 7 presents numerical algorithms used to solve the advection equation describing transport by resolved winds. The formulation of small-scale (parameterized) transport processes including turbulent mixing, organized convection, plumes, and boundary layer dynamics is addressed in Chapter 8. Chapter 9 reviews formulations of emissions to the atmosphere, deposition to the surface, and two-way coupling between the atmosphere and surface reservoirs.

The third part of this book deals with the role of models as components of the atmospheric observing system. Chapter 10 focuses on model evaluation and presents different metrics for this purpose. It illustrates the importance of models for the interpretation of observational data. Chapter 11 covers fundamental concepts of inverse modeling and data assimilation. It shows how chemical transport models can be integrated with atmospheric observations through optimization theory to provide best estimates of the chemical state of the system and of the driving variables.

At the end of the volume, the reader will find several appendices with numerical values of physical constants and other quantities, unit conversions, and a list of important chemical reactions with corresponding rate constants. Some basic mathematical definitions and relations are also provided.

Over the years, both of us have benefited from numerous discussions with our colleagues, students, and postdoctoral fellows. Several of them have contributed to this book by reviewing chapters, making suggestions, and providing scientific material. We are deeply indebted to them. We would like to thank in particular Helen Amos, Alexander Archibald, Jerome Barre, Mary Barth, Cathy Clerbaux, Jim Crawford, Louisa Emmons, Rolando Garcia, Paul Ginoux, Claire Granier, Alex Guenther, Colette Heald, Jan Kazil, Patrick Kim, Douglas Kinnison, Monika Kopacz, Jean-François Lamarque, Peter Lauritzen, Sasha Madronich, Daniel Marsh, Iain Murray, Vincent-Henri Peuch, Philip Rasch, Brian Ridley, Anne Smith, Piotr Smolarkiewicz, Alex Turner, Xuexi Tie, Stacy Walters, Kevin Wecht, Christine Wiedinmyer, Lin Zhang, and Peter Zoogman. We would like also to acknowledge Sebastian Eastham, Emilie Ehretsmann, Natasha Goss, Lu Hu, Rajesh Kumar, Eloise

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Symbols

The symbols used in the different chapters of this book are listed below with their corresponding units in the MKSA system. When no units are given, the quantity is either dimensionless or has no intrinsic dimensions. Appendix B gives further information on units, prefixes, and conversion factors. In some cases, when no confusion exists, the same symbols are used to characterize different variables. Scalars are represented as italics (alphabet letters) or as regular font (Greek and other symbols). Vectors and matrices are represented by lowercase and uppercase bold fonts, respectively.

A

a	Earth's radius [m]
A	Surface area density of atmospheric particles [m^2 m^{-3}]
$\mathbf{A}$	Averaging kernel matrix

B

B	Blackbody radiative emission flux [W m^{-2}]
B_λ	Spectral density of blackbody emission flux (Planck function) [W m^{-2} nm^{-1}]

C

c	One-dimensional constant flow velocity [m s^{-1}]
c	Speed of light in vacuum [m s^{-1}]
c^*	Phase velocity of a wave [m s^{-1}]
c_g^*	Group velocity of a wave [m s^{-1}]
c_p	Specific heat at constant pressure [J K^{-1} kg^{-1}]
c_v	Specific heat at constant volume [J K^{-1} kg^{-1}]
C_c	Slip correction factor
C_D	Drag coefficient
C_i	Mole fraction or molar mixing ratio of species i
$CRMSE$	Centered root-mean-square-error

D

d	Displacement height [m]
D	Divergence of the flow [s^{-1}]
Da	Damköhler number
D_d	Detrainment rate associated with downdrafts in convective systems [kg m^{-3} s^{-1}]
D_i	Molecular diffusion coefficient for species i [m^2 s^{-1}]
D_p	Particle diameter [m]

D_u	Detrainment rate associated with updrafts in convective systems [kg m^{-3} s^{-1}]
$DOFS$	Degrees of freedom for signal

E

e	Water vapor partial pressure [Pa]
e_s	Saturation water vapor pressure [Pa]
$\mathbf{e}$	Eigenvector
E	Emission flux [kg m^{-2} s^{-1}]
$\mathbf{E}$	Eliassen–Palm Flux [components E_φ and E_z in kg s^{-2}]
$\mathbf{E}$	Matrix of eigenvectors arranged by columns
$E(k)$	Spectral distribution of turbulent energy for a given wavenumber k [m^3 s^{-2}]
E_a	Activation energy [J mol^{-1}]
E_d	Entrainment rate associated with downdraft in convective systems [kg m^{-3} s^{-1}]
E_u	Entrainment rate associated with updraft in convective systems [kg m^{-3} s^{-1}]

F

f	Coriolis factor [s^{-1}]
f_A	Fractional area of a model grid cell experiencing precipitation
f_A	Fractional area of land suitable for saltation
$f_{i,I}$	Fraction of soluble compound i partitioned in ice water
$f_{i,L}$	Fraction of soluble compound i partitioned in liquid water
F	Mass flux [kg m^{-2} s^{-1}]
F	Radiative flux [W m^{-2}]
$\mathcal{F}$	Air mass factor
$\mathbf{F}$	Force vector with its three components F_x, F_y, and F_z [N]
$\mathbf{F}$	Forward model
$F_{D,i}$	Deposition flux of species i [kg m^{-2} s^{-1}]
F_λ	Spectral density of the radiative flux [W m^{-2} nm^{-1}]

G

$\boldsymbol{g}$	Vector of gravitational acceleration [m s^{-2}]
g	Amplitude of gravitational acceleration [m s^{-2}]
g	Amplification function in numerical methods
g	Asymmetry factor
g	Gain factor
G	Green function
G	Gravity wave drag [m s^{-2}]
$\mathbf{G}$	Gain matrix
G_m	Grade of model m

H

h	Mixing depth [m]
H	Atmospheric scale height [m]
$\mathcal{H}$	Effective (constant) scale height [m]
H_i	Dimensionless Henry's law constant for species i

I

i	Unit vector in the zonal (x) direction
I	Light intensity [W m^{-2}]
I	Identity matrix
I_{AB}	Segregation ratio for chemical compounds A and B
I_i	Condensation growth rate of species i [m^3 s^{-1}]

J

j	Unit vector in the meridional (y) direction
j	Radiative source term [Wm^{-2} sr^{-1} nm^{-1} m^{-1}]
J	Radiative source function [Wm^{-2} sr^{-1} nm^{-1}]
J	Photodissociation (photolysis) frequency [s^{-1}]
J	Cost function
J	Jacobian matrix
$J_{i,j}$	Coagulation rate between particles i and j [m^{-3} s^{-1}]
J_0	Nucleation rate [m^{-3} s^{-1}]

K

k	Unit vector in the vertical (z) direction
k	Wavenumber [m^{-1}]
k	Boltzmann's constant (1.38 × 10^{-23} J K^{-1})
k	von Karman's constant (0.35)
k	Chemical rate constant [first order: s^{-1}; second order: cm^3 s^{-1}; third order: cm^6 s^{-2}]
k_{ext}	Mass extinction cross-section [m^2 kg^{-1}]
$k_{G,i}$	Conductance for vertical transfer of species i in the gas phase [m s^{-1}]
$k_{W,i}$	Conductance for vertical transfer of species i in the water phase [m s^{-1}]
K	Eddy diffusion coefficient [m^2 s^{-1}]
K	Equilibrium constant
K	Henry's law constant [M atm^{-1}]
K^*	Effective Henry's law constant [M atm^{-1}]
K	Eddy diffusion tensor
K	Jacobian matrix (Chapter 11)
K_a	Acid dissociation constant
K_m	Eddy viscosity coefficient [m^2 s^{-1}]
Kn	Knudsen number
K_i	Air–sea exchange velocity for species i [m s^{-1}]
K_θ	Eddy diffusivity of heat [m^2 s^{-1}]

L

l	Mixing length [m]
ℓ_i	Loss rate constant or loss coefficient of species i [s^{-1}]
L	Characteristic length [m]
L	Liquid water content [kg water/kg air]
L	Monin–Obukhov length [m]

L	Lagrange function
L_i	Loss rate of species i [m^{-3} s^{-1}]
L_{vap}	Latent heat of vaporization of liquid water [J kg^{-1}]
L_λ	Spectral density of the radiance at wavelength λ [W m^{-2} sr^{-1} nm^{-1}]

M

m	Mean molecular mass of air (4.81 × 10^{-26} kg)
m	Refraction index
m	Wavenumber
M_a	Molar mass of air (28.97 × 10^{-3} kg mol^{-1})
M_d	Mean vertical downdraft convective flux of air [kg m^{-2} s^{-1}]
M_e	Mean subsidence flux compensating for convective fluxes [kg m^{-2} s^{-1}]
M_i	Molar mass of species i [kg mol^{-1}]
M_k	Moment of order k for a given aerosol distribution
M_u	Mean vertical updraft convective flux of air [kg m^{-2} s^{-1}]
M_w	Molar mass of water (18.01 × 10^{-3} kg mol^{-1})
MAD	Mean absolute deviation
MAE	Mean absolute error
MFB	Mean fractional bias
MFE	Mean fractional error
$MNAE$	Mean normalized absolute error
MNB	Mean normalized bias

N

$\mathbf{n}$	Unit outward vector normal to a surface
n_a	Number density for air [m^{-3}]
n_i	Number density for species i [m^{-3}]
n_N	Particle number size distribution function [m^{-4}]
n_S	Particle surface distribution function [m^2 m^{-4}]
n_V	Particle volume distribution function [m^3 m^{-4}]
$\mathcal{N}_A$	Avogadro number (6.022 × 10^{23} molecules per mole)
NMB	Normalized mean bias

P

p	Pressure [Pa]
p_d	Pressure of dry air [Pa]
p_i	Production rate of species i [kg m^{-3} s^{-1}]
p_s	Surface pressure [Pa]
P	Phase function for scattered radiation
P	Ertel potential vorticity [m^2 s^{-2} K kg^{-1}]
P	Probability density function
$\mathcal{P}$	Steric factor
Pe	Péclet number
P_i	Production rate of species i [m^{-3} s^{-1}]
P_l^m	Associated Legendre polynomial
Pr	Prandtl number

Q

q	Specific humidity [kg water vapor/kg of air]
q	Diabatic heating expressed in K day^{-1}
q	Actinic flux [photons m^{-2} s^{-1}]
q_k	Water concentration in hydrometeor of type k
q_λ	Photon flux density [photons m^{-2} s^{-1} nm^{-1}]
Q	Diabatic heating rate [J kg^{-1} s^{-1} or W m^{-3}]
Q_{abs}	Absorption efficiency
Q_{ext}	Extinction efficiency
Q_s	Saltation flux [kg m^{-1} s^{-1}]
Q_{scat}	Scattering efficiency

R

r	Geometric distance from the center of the Earth
$\mathbf{r}$	Position vector
r	Particle radius [m]
r_w	Mass mixing ratio of water vapor [kg kg^{-1}]
r	Pearson correlation coefficient
R	Gas constant for air [J K^{-1} kg^{-1}]
$\mathcal{R}$	Universal gas constant (8.3143 J K^{-1} mol^{-1})
R^2	Coefficient of determination
R_A	Aerodynamic resistance [s m^{-1}]
$R_{B,i}$	Boundary resistance for species i [s m^{-1}]
$R_{C,i}$	Surface resistance for species i [s m^{-1}]
R_d	Gas constant for dry air (287 J K^{-1} kg^{-1})
Re	Reynolds number
RH	Relative humidity [percent]
Ri	Richardson number
R_i	Total resistance to dry deposition of species i [s m^{-1}]
$RMSE$	Root mean square error
R_w	Gas constant for water vapor (461.5 J K^{-1} kg^{-1})

S

s_i	Source rate of species i (in mass) [kg m^{-3} s^{-1}]
S	Solar energy flux [W m^{-2}] or solar constant (approx. 1368 W m^{-2})
$\mathbf{S}$	Error covariance matrix
$\mathbf{S}'$	Error correlation matrix
$\mathbf{S_a}$	Aggregation error covariance matrix
$\mathbf{S_A}$	Prior error covariance matrix
$\mathbf{S_I}$	Instrument error covariance matrix
$\mathbf{S_M}$	Forward model error covariance matrix
$\mathbf{S_O}$	Observational error covariance matrix
$\mathbf{S_R}$	Representation error covariance matrix
$\hat{\mathbf{S}}$	Posterior error covariance matrix
Sc_i	Schmidt number for species i

T

t	Time [s]
t	Student's variable for the *t*-test
T	Transmission of radiation
T	Absolute temperature [K]
T_E	Effective temperature of the Earth [K]
TKE	Turbulent kinetic energy [m^2 s^{-1}]
T_s	Effective temperature of the Sun
T_v	Virtual temperature [K]

U

u	Zonal component of wind velocity [m s^{-1}]
u	Path length [kg m^{-2}]
u_*	Friction velocity [m s^{-1}]
u^*	Residual zonal wind velocity [m s^{-1}]
u^A	Anti-diffusion velocity [m s^{-1}]
u_g	Zonal component of the geostrophic wind [m s^{-1}]
u_{10}	Wind velocity 10 m above the surface [m s^{-1}]

V

v	Meridional component of wind velocity [m s^{-1}]
v^*	Residual meridional wind velocity [m s^{-1}]
$\mathbf{v}$	Wind velocity vector in Earth's rotating frame [m s^{-1}]
v_g	Meridional component of the geostrophic wind [m s^{-1}]
v_i	Mean thermal velocity [m s^{-1}]
V	Molar volume [m^3 mol^{-1}]
V	Aerosol volume density [m^3 m^{-3}]
$\mathbf{V}$	Wind velocity in inertial frame [m s^{-1}]
V_T	Translational Earth's rotation velocity [m s^{-1}]

W

w	Vertical component of wind velocity [m s^{-1}]
$w*$	Residual vertical wind velocity [m s^{-1}]
$w*$	Convective velocity scale [m s^{-1}]
$w_{D,i}$	Surface deposition velocity of species *i* [m s^{-1}]
w_s	Terminal settling velocity [m s^{-1}]

X

x	Geometric distance in the zonal direction [m]
$\mathbf{x}$	State vector (often refers to the true value)
$\hat{\mathbf{x}}$	Optimal estimate of state vector
$\mathbf{x_A}$	Prior estimate of state vector

Y

y	Geometric distance in the meridional direction [m]
$\mathbf{y}$	Observation vector

Z

z	Geometric altitude [m]
$z_{0,m}$	Aerodynamic roughness length [m]
Z	Log pressure altitude [m]
Z	Potential vorticity [s^{-1} m^{-1}]
Z_{AB}	Collision frequency for molecules A and B [s^{-1}]

α

α	Albedo
α	Aerosol particle size parameter
α	Mass accommodation coefficient
α	Courant number
α_T	Thermal diffusion factor

β

β	Fourier number
β_{ext}	Aerosol extinction coefficient [m^{-1}]
β_{abs}	Aerosol absorption coefficient [m^{-1}]
β_{scat}	Aerosol scattering coefficient [m^{-1}]
$\beta_{i,j}$	Coagulation coefficient for particles i and j [m^3 s^{-1}]

γ

γ	Reactive uptake coefficient for heterogeneous chemical process
γ	Regularization factor
γ_c	Coefficient for non-local turbulent transfer
Γ	Actual atmospheric lapse rate [K m^{-1}]
Γ	Mean age of air [s]
Γ_d	Dry adiabatic lapse rate [K m^{-1}]
Γ_w	Wet adiabatic lapse rate [K m^{-1}]
$\boldsymbol{\Gamma_{\varpi}}$	Aggregation matrix

δ

δ	Dirac function
ΔH	Enthalpy of dissolution [J mol^{-1}]

ε

ε_A	Quantum efficiency (or yield) for the photolysis of molecule A
$\boldsymbol{\varepsilon_O}$	Observational error vector
$\boldsymbol{\varepsilon_a}$	Aggregation error vector
$\boldsymbol{\varepsilon_A}$	Prior estimate error vector
$\boldsymbol{\varepsilon_I}$	Instrument error vector
$\boldsymbol{\varepsilon_M}$	Forward model error vector
$\boldsymbol{\varepsilon_R}$	Representation error vector

ζ

ζ	Relative vorticity of the flow [s^{-1}]

η

η	Step mountain coordinate (eta coordinate)

θ

θ	Zenithal direction [radians]
θ	Potential temperature [K]
θ_v	Virtual potential temperature [K]

λ

λ	Longitude [radians]
λ	Wavelength [m]
λ	Mean free path of air molecules [m]
λ	Lyapunov exponent [s^{-1}]
λ_i	Eigenvalue associated with eigenvector $\mathbf{e}_i$
Λ	Leaf area index (LAI) [$m^2\ m^{-2}$]

μ

μ	Cosine of zenithal direction (θ)
μ	Molecular dynamic viscosity coefficient [Pa s or kg $m^{-1}\ s^{-1}$]
μ_i	Mass mixing ratio of species *i* [kg kg^{-1}]
μ_w	Mass mixing ratio of water vapor [kg kg^{-1}]

ν

ν	Kinematic viscosity [$m^2\ s^{-1}$]
ν	Asselin-filter parameter
ν	Frequency [Hz]
ν_{ion}	Ion-neutral collision frequency [s^{-1}]

π

π	3.14159

ρ

ρ_a	Mass density of air [kg m^{-3}]
ρ_d	Mass density of dry air [kg m^{-3}]
ρ_i	Mass density of species *i* [kg m^{-3}]
ρ_p	Mass density of particles or drops [kg m^{-3}]
ρ_w	Mass density of water vapor [kg m^{-3}]

σ

σ	Stefan-Boltzmann constant (5.67×10^{-8} W m^{-2} K^{-4})
σ	Standard deviation
σ	Normalized pressure coordinate (sigma coordinate)
$\tilde{\sigma}$	Pseudo density in isentropic coordinates
σ_A	Absorption cross-section for molecule A [m^2]

τ

τ	Optical depth
τ	Lifetime [s]
$\boldsymbol{\tau}$	Stress tensor
$\tau_{i,j}$	Element of the stress tensor

φ

φ	Latitude [radians]
φ	Azimuthal direction
ϕ	Radial basis function
Φ	Geopotential [$m^2\ s^{-2}$]
$\boldsymbol{\Phi}_\infty$	Solar flux at the top of the atmosphere [$W\ m^{-2}$]
Φ_k	Basis function in the spectral element method
Φ_λ	Spectral density of solar flux [$W\ m^{-2}\ nm^{-1}$]

χ

χ	Solar zenith angle
χ	Velocity potential

ψ

Ψ	Generic mathematical function or variable
Ψ	Streamfunction of the flow
Ψ	Montgomery function (isentropic coordinate system) [$J\ kg^{-1}$ or $m^2\ s^{-2}$]

ω

ω	"Vertical" velocity in the pressure coordinate system [$Pa\ s^{-1}$]
ω	Single scattering albedo
Ω	Angular Earth rotation period (7.292×10^{-5} rad s^{-1})
Ω	Column concentration [molecules m^{-2}]
Ω_s	Slant column concentration [molecules m^{-2}]