

Guide

Guide to Global Aerosol Models (GAM)

AIAA standards are copyrighted by the American Institute of Aeronautics and Astronautics (AIAA), 1801 Alexander Bell Drive, Reston, VA 20191-4344 USA. All rights reserved.

AIAA grants you a license as follows: The right to download an electronic file of this AIAA standard for temporary storage on one computer for purposes of viewing, and/or printing one copy of the AIAA standard for individual use. Neither the electronic file nor the hard copy print may be reproduced in any way. In addition, the electronic file may not be distributed elsewhere over computer networks or otherwise. The hard copy print may only be distributed to other employees for their internal use within your organization.

Guide to Global Aerosol Models

Sponsor

American Institute of Aeronautics and Astronautics

Abstract

This Guide defines a set of existing reference global aerosol models. It includes tables, plots, and other information describing the optical and mechanical properties of atmospheric aerosols, both natural and man-made. Intended for use by the aerospace and meteorological communities, the emphasis is on the physical and chemical characteristics, size distributions, sources, locations, and sinks. Urban, rural, maritime, desert, and polar locations; combustion and volcanic sources; and geographic variations are all treated.

Guide to global aerosol models/sponsor, American Institute of
Aeronautics and Astronautics.

p. cm.

"AIAA G-065-1999"

Includes bibliographical references.

ISBN 1-56347-348-8 (softcover), 1-56347-369-0 (electronic)

1. I. American Institute of Aeronautics and

Astronautics.

QC882.42.G85 1999

551.51' 13—dc21

99-36696
CIP

Published by

American Institute of Aeronautics and Astronautics
1801 Alexander Bell Drive, Reston, VA 22091

Copyright © 1999 American Institute of Aeronautics and Astronautics
All rights reserved

No part of this publication may be reproduced in any form, in an electronic
retrieval system or otherwise, without prior written permission of the publisher.

Printed in the United States of America

Contents

	Page
List of Figures	iv
List of Tables	vi
Foreword.....	vii
1 Introduction	1
1.1 Scope	1
1.2 Aerosol Background.....	1
1.3 Sources for Raw Data	1
1.4 History	2
1.5 Structure of the Guide	3
2 Global Aerosol Characteristics.....	4
2.1 Sources, Production Rates, and Vertical Distribution	4
2.2 Aerosol Size Distribution	5
2.2.1 Characterization of Atmospheric Aerosols.....	5
2.2.2 Aerosol Size Distribution Functions.....	5
2.3 Aerosol Types.....	9
2.4 Aerosol Variations.....	12
2.5 Aerosol Optical Properties.....	15
2.5.1 Optical Properties of a Single Particle	16
2.5.2 Optical Properties of a Cloud of Particles	17
2.5.3 Aerosol Refractive Indices.....	19
2.6 Aerosol Mechanical Characteristics.....	22
3 Global Aerosol Models.....	23
3.1 Boundary Layer	24
3.1.1 General Models	24
3.1.2 Urban Boundary Layer.....	25
3.1.3 Rural Boundary Layer.....	25
3.1.4 Maritime Boundary Layer.....	25
3.1.5 Desert Boundary Layer.....	28
3.1.6 Polar Boundary Layer.....	29
3.1.7 Combustion Products and Their Transport.....	29
3.1.8 Optical Characteristics.....	31
3.2 Free Troposphere.....	35
3.2.1 Vertical Aerosol Distribution	35
3.2.2 Continental and Maritime Background	37
3.2.3 Desert Dust Streams	42
3.2.4 Combustion Products	42
3.2.5 Arctic Haze.....	42
3.2.6 Volcanic Contamination, Stratospheric/Tropospheric Exchange	42
3.3 Stratosphere and Mesosphere.....	43
3.3.1 Introduction	43
3.3.2 Intervolcanic Background.....	44
3.3.3 Volcanic	52
3.3.4 Polar Stratospheric Clouds	56
3.3.5 Mesospheric Clouds	58
3.3.6 HSST	59
Annex A: The Altitude-Size Distribution.....	60
References.....	61

List of Figures

	Page
Figure 1.	Characteristics of the atmospheric aerosols as a function of altitude 5
Figure 2.	Normalized frequency plots of the number, surface, and volume distributions as a function of particle size for the grand average 1969 Pasadena smog aerosol 8
Figure 3.	Atmospheric aerosol surface area distribution showing the principal modes, the main source of mass in each mode, and the principal removal mechanisms. 9
Figure 4.	Mass concentration of mineral aerosols (in $\mu\text{g m}^{-3}$) for April at the 1000 hPa pressure level 12
Figure 5.	Aerosol modification and removal processes..... 13
Figure 6.	Aerosol residence time of individual particles as a function of particle size and altitude in the atmosphere..... 13
Figure 7.	Scattering by a single particle at p..... 16
Figure 8.	Mie scattering diagram..... 17
Figure 9.	Dependence of the efficiency factor Q_e on the size parameter. 17
Figure 10.	Scattering by an element dV of the aerosol medium of total volume V 18
Figure 11.	Transmission law for an aerosol medium of length L 19
Figure 12.	Schematic showing the various geographical and altitude regions used in defining aerosol models. 23
Figure 13.	Model number size distributions of selected atmospheric aerosols according to Table 10 25
Figure 14.	Model dry salt aerosol distributions (shown as area) developed from the combination of OPC and Woodcock size distributions..... 26
Figure 15.	Equilibrium aerosol diameters as a function of relative humidity for various aerosol compositions 28
Figure 16.	Variation of (a) desert aerosol particle number density and (b) surface area with wind velocity 29
Figure 17.	The differential spectra of the areas of continental aerosol particles 29
Figure 18(a).	Size distributions of various kinds of particles collected following the Kuwait oil fires 31
Figure 18(b).	Size distributions of aerosol samples collected at various distances from the fire but at similar altitudes..... 31
Figure 19.	Relationship between black carbon concentrations and distance from China shore 32
Figure 20.	Radiatively equivalent aerosol optical thickness (EAOT x 1000) over the oceans derived from NOAA AVHRR satellites for the four seasons..... 33
Figure 21.	Seasonal variation of regional optical depth 34
Figure 22.	Mean values and variabilities of the extinction and absorption coefficients of dry atmospheric particles at different air masses..... 35
Figure 23(a).	Altitude distributions of particle mass concentration..... 36
Figure 23(b).	Altitude distribution of aerosol number concentration..... 36
Figure 24.	Vertical distribution of dust concentration for the four different size classes (averaged over all longitudes) in units of microgram dust per kilogram air 38
Figure 25.	Long-term average bimodal log normal tropospheric aerosol particle size distributions at Laramie for two altitude intervals during summer (left) and winter (right)..... 39
Figure 26.	Elemental composition of aerosols over the Pacific Basin as a function of altitude..... 41
Figure 27.	Seasonal variations in zonally averaged background aerosol characteristics at an altitude of 7.5 km 41
Figure 28.	Log-normal cubic spline model size distribution for two extreme conditions at the peak of the stratospheric aerosol layer 44

List of Figures (continued)

	Page
Figure 29. Global-averaged, annual mean profiles of 1.0-micrometer aerosol extinction from the 1979 and 1989 reference models.....	45
Figure 30. Time-latitude section of stratospheric aerosol optical depth at 1 μm , blending SAGE I and II and SAM II data	48
Figure 31. Aerosol surface area density in square microns per cubic centimeter	48
Figure 32. Background mass mixing ratio as a function of altitude.....	49
Figure 33. Aerosol surface area density as a function of time and latitude.....	50
Figure 34. Estimated stratospheric aerosol optical depth	52
Figure 35. Time series from SAM II of weekly averaged values of aerosol optical depth at high latitudes.....	55
Figure 36. Time series of one degree zonally averaged optical thickness departures starting at the time just after the Mt. Pinatubo eruption	56
Figure 37(a). Seasonal variation in the averaged frequency of occurrence of Arctic PSC sightings by SAM II for the period 1978–89.....	57
Figure 37(b). Seasonal variation in the averaged frequency of occurrence of Antarctic PSC sightings by SAM II for the period 1978–89.....	57
Figure 38(a). Histogram showing the distribution with longitude of Arctic PSC sightings by SAM II for the period 1978–89	58
Figure 38(b). Histogram showing the distribution with longitude of Antarctic PSC sightings by SAM II for the period 1978–89	58

List of Tables

	Page
Table 1. Commonly Used Techniques for Measuring Aerosol Characteristics	2
Table 2. Important General References on Aerosol Models and Characteristics.....	3
Table 3. Source Strength of Atmospheric Aerosol Particles in Tg/yr for Radii Smaller than 100 μm	4
Table 4. Commonly Used Size Distribution Functions.....	7
Table 5. Aerosol Types	10
Table 6. Aerosol Components and Contribution to Aerosol Types.....	11
Table 7. Aerosol Equivalent Volume Radius at Different Relative Humidities.....	15
Table 8. Complex Refractive Index ($m_r + im_i$) of Various Aerosol Components and Water.....	20
Table 9(a). Moh's Scale-of-Hardness for Minerals	22
Table 9(b). Moh's Hardness Values for Some Other Minerals.....	22
Table 10. Parameters for Models of Aerosol Size Distributions Described by the Sum of Three Log Normal Functions	24
Table 11. Log Normal Parameters for Sea Salt Aerosol Number Size Distributions ($dN/d\log D$).....	27
Table 12. Wind Speed Dependence of the Desert Aerosol Size Distribution Parameters.....	28
Table 13. Parameters of Vertical Aerosol Profiles	36
Table 14. Vertical Distribution of Atmospheric Aerosol in 3 or 4 Layers	37
Table 15. Tropospheric Aerosol Mean Seasonal Size Distribution and Mass	40
Table 16. Free Tropospheric Size Distribution Parameter for the Pacific Basin	40
Table 17. Annually Smoothed Mass Loading	42
Table 18. Tropospheric Volcanic Data Intercomparisons	42
Table 19. Characteristics of Stratospheric Aerosols	43
Table 20. Parameters for Size Distribution.....	45
Table 21(a). Coefficients of the Polynomial Derived from SAGE I 1.0-Micrometer Aerosol Extinction (March 1979 to February 1980).....	46
Table 21(b). Coefficients of the Polynomial Derived from SAGE II 1.0-Micrometer Aerosol Extinction (March 1989 to February 1990) (Concluded).....	46
Table 22. Decadal Mean Values of Aerosol Extinction at 1 μm , in Units of 10^{-4}km^{-1} , Given Every 10° From 80°S to 80°N and Every 2 km from 8 km (or the Tropopause) to 34 km Altitude.....	47
Table 23. Estimated Annual Stratospheric Aerosol Optical Depths (Multiplied by 1000) at $\lambda = 0.55 \mu\text{m}$	53
Table 24. Estimate of Aerosol Mass Loading in the Stratosphere.....	55

Foreword

This *Guide to Global Aerosol Models* has been sponsored by the American Institute of Aeronautics and Astronautics (AIAA) as part of its Standards Program.

The purpose of this Guide is to provide, in convenient form, a set of reference global aerosol models (GAM) for use by the aerospace community. Information is provided on aerosol physical and chemical characteristics, size distribution, physical state, and mass concentration, as well as on their basic optical properties.

The scope of the effort is confined to aerosols commonly found in the atmosphere, particularly those arising from natural causes; somewhat less emphasis is placed on aerosols produced by human development and industry. The aim has been to provide a general survey of aerosol types and characteristics, rather than detailed physical and chemical properties. Derived quantities, such as atmospheric radiative characteristics, have similarly been omitted.

The information provided in the form of tables and graphs is taken from published sources. No attempt has been made to create new data or new models. In this effort, assistance was sought from a worldwide community of aerosol scientists. A draft outline of the guide was prepared and distributed to over 40 atmospheric aerosol scientists around the world. The following individuals, who responded by sending and/or suggesting relevant literature, tables, and models, comprise the AIAA Global Aerosol Model Working Group. They are

A. Deepak, Chairman (Science and Technology Corporation, USA)
 A. D. Clarke (University of Hawaii, USA);
 D. A. Gillette (National Oceanic and Atmospheric Administration (NOAA), USA);
 G. Hänel (Johann Wolfgang Goethe-Universität, Germany);
 P. V. Hobbs (University of Washington, USA);
 R. Jaenicke (Johannes Gutenberg-Universität, Germany);
 J. Lenoble (Université des Sciences et Technologies de Lille, France);
 G. S. Kent (Science and Technology Corporation, USA)
 F. Parungo (Science and Technology Corporation, USA);
 J. Podzimek (University of Missouri-Rolla, USA);
 E. P. Shettle (Naval Research Laboratory, USA);
 K. S. Shifrin (Oregon State University—Corvallis, USA);
 L. L. Stowe (NOAA, USA).

Their assistance in this effort is gratefully acknowledged.

To facilitate convenient access to aerosol characteristics and models, this guide is structured so that the early sections provide general information on aerosol properties and classification, as well as on the nomenclature used to describe them. The latter sections contain more detailed models and data relating to aerosol normally occurring in specific atmospheric regions and environments.

It is a pleasure to acknowledge the assistance of STC staff, especially M. D. (Sue) Crotts, Linda Schofield, and Hope Tarr, in the preparation of the camera-ready copy of this guide. It is hoped this volume will be a valuable contribution to the international standards on global aerosol models for the aerospace community, and will act as a useful reference point for future updates.

The AIAA Standards Procedures provide that all approved Standards, Recommended Practices, and Guides are advisory only. Their use by anyone engaged in industry or trade is entirely voluntary. There is no agreement to adhere to any AIAA standards publication and no commitment to conform to or be guided by any standards report. In formulating, revising, and approving standards publications, the Committees on Standards will not consider patents which may apply to the subject matter. Prospective users of the publications are responsible for protecting themselves against liability for infringement of patents or copyrights, or both.

The AIAA Atmospheric & Space Environments Committee on Standards (Shu T. Lai, Chairman) approved the document in January 2000.

The members of this consensus body at the time of voting on the document are:

Dana A. Brewer (NASA Headquarters)
Herbert C Carlson (Air Force Office of Scientific Research)
Adarsh Deepak (Science & Technology Corp.)
Gerald Dittberner (NOAA/NEDIS)
L. J. Ehernberger (NASA Ames-Dryden Facility)
Jeffrey M. Forbes (University of Colorado)
Henry B. Garrett (Jet Propulsion Lab.)
G. Barry Hillard (NASA Lewis Research Center)
Stuart L. Huston (Boeing Space & Defense)
JoAnn Joselyn (NOAA Space Environment Lab.)
Neil D. Kelley (National Renewable Energy Lab.)
O. Kenneth Moe (USAF Space & Missile Systems Center)
Jerry Owens (NASA Marshall Space Flight Center)
Robert Sears (Jamieson Science & Engineering)
Robert A. Skrivanek (Visidyne Inc.)
Guy F. Spitale (Jet Propulsion Laboratory)
Walther Spjeldvik (Weber State University)
Robert M. Suggs (NASA Marshall Space Flight Center)
Gopal D. Tejwani (Lockheed Martin Space Operations.)
Alfred Vampola (Consultant)
William W. Vaughan (UAH Research Center)

The AIAA Standards Executive Council accepted the document for publication in January 2000

1 Introduction

1.1 Scope

The purpose of this work is to describe, in a convenient form, a standard Global Atmospheric Model which is composed of a set of existing reference aerosol models. This guide contains tables, plots, and other information describing the properties of atmospheric aerosols, both natural and man-made. These are intended for use by the aerospace community and the emphasis is on the physical and chemical characteristics, size distributions, sources, locations, and sinks. Somewhat less emphasis is placed upon aerosol optical properties, which are discussed in Section 2.5.

Information provided is based on published sources, particularly models and summary data. No attempt has been made to create new models or tables directly from measured data.

1.2 Aerosol Background

An “aerosol” is a suspension of finely divided matter (liquid or solid) dispersed in a gaseous medium such as air. The suspended particles are called “aerosol particles” or “aerosols.”

Aerosol particles which occur naturally in the atmosphere are called “atmospheric aerosols;” those which are produced in the laboratory or field space by man-made chemical, thermal, or mechanical means are called “artificial aerosols.” In this work, the term atmospheric aerosols will cover aerosols derived from both natural and anthropogenic sources, the latter being those generated as by-products of human activity, such as burning of fossil fuels and are often referred to as “anthropogenic aerosols.”

Atmospheric aerosols may be formed as the result of wind action at the earth’s surface (land or ocean) or formed *in situ* by condensation from the gaseous phase. Gaseous precursors may, in turn, be formed by the action of sunlight and chemical action on other naturally or anthropogenically-produced gases. Aerosols are subject to changes in size and composition by processes of condensation, evaporation, and coagulation. Water soluble aerosols may also change size in response to changes in atmospheric humidity. Aerosols, particularly those in the troposphere, are often mixtures of different chemical components. Such mixtures may be external, in which different particles within the same air mass have differing compositions, or “internal,” in which individual particles are of mixed composition. An example of a relatively simple homogeneous aerosol is that found in the stratosphere which, except just after a volcanic eruption, consists of a solution of sulfuric acid in water that is in equilibrium with its surroundings. The marine boundary layer aerosol is an example of an external mixture, in which aerosols of different compositions (sea salt and sulfates) occupy different parts of the size distribution spectrum. Internal mixtures are often formed within the atmosphere, as for example, when a solid desert aerosol passes through an industrial plume and the individual particles acquire a layer of sulfuric acid. Atmospheric aerosols are also subject to modification by cloud processes, where aerosols becomes integrated into the cloud droplets and may be later regenerated, often in modified form, as a result of evaporation. Cloud processes are also responsible for the removal of much of the aerosol from the atmosphere within rain or snow.

Aerosol concentrations and compositions depend on location relative to source regions, production and removal rates, transport, and altitude. It is possible to classify aerosols according to their sources and to describe their approximate chemical composition and size distribution. The most significant sources for atmospheric aerosols lie within the planetary boundary layer, and it is here that the largest concentrations are normally to be found. Sources are not only localized but may also vary in time. These variations may be seasonal, such as in the formation of desert dust plumes by spring-time surface wind erosion or in the annual cycle of biomass burning and smoke production in tropical countries. Examples of sporadic aerosol production are those formed as a result of volcanic activity or forest fires.

1.3 Sources for Raw Data

Numerous data sources exist for model development. Data may be obtained by *in situ* or remote techniques. Measurements of size distributions, chemical composition, and the characteristics of individual particles normally require *in situ* instruments. Most commonly, these instruments will be located at the earth’s surface but airborne and, somewhat less commonly, balloon-borne measurements are also made. The flexibility of airborne measurements lies not only in the ability to sample different altitudes, but also in its capability to reach remote locations.

Bulk aerosol properties may be measured by both *in situ* and remote techniques. Remote methods are typically optical and include lidar and satellite techniques, the latter giving global coverage but requiring careful validation.

and interpretation for proper understanding of the data. *In situ* measurements are also often, but not uniquely, optical. Particle collection, plus mass measurement and chemical analysis play an important role. A list of commonly used techniques for measuring the characteristics and behavior of atmospheric aerosols is given in Table 1 and is taken from Stowe et al.

Table 1. Commonly Used Techniques for Measuring Aerosol Characteristics from Space, Airborne, and Ground-Based Platforms

Remote Sensors		In situ Sensors
Lidars	(s,a,g)	Optical Spectrometers
Transmissometers	(g)	Impactors
Photometers (λ)	(a,g)	Electric Mobility Analyzers
Radiometers (λ)	(s,a,g)	Diffusion Mobility Analyzers
Multi-Spectral	(s,a,g)	Filter Samplers
Shadowband	(g)	Condensation Nuclei Counters
Solar Occultation	(s)	Ice Nuclei Counters
Limb Scattering	(s)	Mass Spectrometers
Surface Radiance	(s)	

Legend: s–space-borne; a–airborne; g–ground-based

1.4 History

Many publications exist that list aerosol models and characteristics. Most of these are specific in nature but several form important reference works that summarize a wide range of aerosol types and characteristics. A short list of these is given in Table 2. They supply much background information to the tables given in the present monograph and in some cases have supplied the tables reproduced here. There is also much overlapping and duplicated information between these sources.

Table 2. Important General References on Aerosol Models and Characteristics

Authors	References	Notes
World Meteorological Organization (WMO)	<i>Aerosols and Climate: Report of the Meeting of Experts on Aerosols and Climate</i> . Geneva, 27–31 October 1980. World Meteorological Organization, WCP-12	Reviews the role of aerosols in climate, aerosol models, measurements, present state of knowledge.
A. Deepak, H.E. Gerber	<u>Report of Experts Meeting on Aerosols and Their Climatic Effects</u> . Williamsburg, VA., 28-30 March 1983, World Meteorological Organization, WCP-55.	Summarizes aerosol physical properties, effects on climate, available data and recommended improvements.
R. Jaenicke	<i>Aerosol Physics and Chemistry in Landolt-Borstein Numerical Data and Functional Relationships in Science and Technology</i> . V, 4b (Meteorology), 1988.	Numerous tables and graphs covering physical, chemical, and optical properties of aerosols. Instrumentation and sampling also described.
R. Jaenicke	<i>Tropospheric Aerosols in Aerosol-Cloud-Climate Interactions</i> . Ed. P.V. Hobbs. Academic Press, 1993.	A review of tropospheric aerosols, types, sources, and behavior. Source of some of the tables used in present monograph.
D'Almeida, P. Koepke, and E.P. Shettle	<i>Atmospheric Aerosols, Global Climatology, and Radiative Characteristics</i> . A. Deepak Publishing, Hampton, VA, 1991.	Aerosol types, sources, characteristics, size distributions, and composition. Emphasis on radiative characteristics.
J. Prospero, R.J. Charlson, V. Mohnen, R. Jaenicke, A.C. Delany, J. Moyers, W. Zoller, and K. Ruhn	The Atmospheric Aerosol System: An Overview. <i>Rev. Geophys. and Space Physics</i> , 21, 1607–1629, 1983.	General article on aerosols and their interactions with the environment.

1.5 Structure of the Guide

This guide consists of three sections. Sections 1 and 2 contain introductory and background material. Section 3 contains the models for individual regions of the atmosphere included in the GAM. Further information on these is given at the beginning of Section 3. Section 2 of this guide gives an overview of global atmosphere aerosol characteristics. In Section 2, we discuss and list aerosol sources, size distributions, and associated analytical models, classification by type, and the component structure that makes up these aerosol types. An overview is also presented of the vertical aerosol structure in the atmosphere, aerosol variations in time and space, modification and removal processes, and dependence upon humidity. Subsections of this section also include descriptions of aerosol optical properties, and how these are characterized, as well as aerosol mechanical properties. Some of the information given in Section 2 may be needed as background for the appreciation of the detailed models given in Section 3.

2 Global Aerosol Characteristics

2.1 Sources, Production Rates, and Vertical Distribution

Atmospheric aerosol sources may be divided into two general classes natural and anthropogenic. Inside these classes, it is possible to make a further subdivision into direct particle production at the earth's surface and formation within the atmosphere from gas to particle conversion and cloud evaporation. Estimates for the annual production rates of atmospheric particles from the various sources are shown in Table 3. These estimates are in order of magnitude only and different authors may give quite different numbers. There are three large natural sources, all producing of the order of 10^9 metric tons of aerosol per year. Salt particles are produced at the oceans' surface as a result of wind action. These have a wide range of sizes, the largest of which are not transported very far. Wind action similarly produces aerosols from the earth's deserts and other arid regions. These may be transported considerable distances away from the source regions. The third important natural source consists of aerosols formed by nucleation and condensation of naturally produced precursor gases. These gases form aerosols containing sulphate and nitrates, as well as organic materials. Less significant natural sources include volcanic debris, natural biologic particles, soot from wild fires, and extraterrestrial dust. A further source of natural aerosols, not included in Table 3 are those produced by evaporation of cloud [Jaenicke, 1993].

Table 3. Source Strength of Atmospheric Aerosol Particles in Tg/yr for Radii Smaller than 100 μm (Data taken from D'Almeida *et al.* [1991] with modifications from Jaenicke [1993])

Aerosol Component	Source Strength
Natural Aerosol Sources	
Extraterrestrial dust	10 - 18
Sea-salt	1,000 - 10,000
Desert dust	500 - 2,000
Volcanic debris	15 - 250
Biogenic/biologic	80
Gas to Particle Conversion (sulfate, organics, nitrate)	345 - 1,300
Subtotal	1925 - 13,648
Man-Made Aerosol Sources	
Direct particulate emission	10 - 90
Gas to Particle Conversion (carbonaceous substances, organics, sulfate, nitrate)	175 - 325
Forest fires and slash-burning debris*	3 - 150
Subtotal	188 - 565
TOTAL	2,673 - 14,213

* Includes unknown amount of indirect natural contributions.

The mass of aerosol produced by man-made activities is considerably less than that produced naturally. As much of this is produced within the atmosphere by gas-to-particle conversion and has a considerable residence time, its regional impact may be out of proportion to its mass. In addition to particles formed from gaseous anthropogenic emissions, others are produced mechanically and as the result of combustion (power production, agricultural burning, and road traffic).

Figure 1 shows a simple schematic of the general character of atmospheric aerosols from the earth's surface to 200-km altitude. Above an altitude of about 30 km, most aerosols are likely to be of extra-terrestrial origin and to exist in low concentrations. A particular phenomenon is the occurrences of noctilucent clouds in certain latitudes at an altitude of about 80 km. Between the tropopause and 30 km, stratospheric aerosols consist of sulfuric acid and are mainly of volcanic origin, formed by oxidation and hydrolysis of SO_2 injected directly into the atmosphere. At

times of low volcanic activity, transformation of stable surface derived gases, such as OCS, into sulphuric acid may be the dominant mechanism in this region. The upper troposphere within a few kilometers of the tropopause represents an overlapping region in which aerosols may be lifted from below or derived from the stratosphere by sedimentation and dynamical processes. In the two lowest regions, aerosols are derived from surface sources. The structure, depth, and aerosol content in the boundary layer is determined by the local sources and meteorological conditions (such as the height of the inversion layer), with aerosol concentration normally decreasing with altitude. Above the boundary layer, surface-derived aerosols have lower concentrations, often exhibit a layered structure and show evidence of considerable horizontal transport.

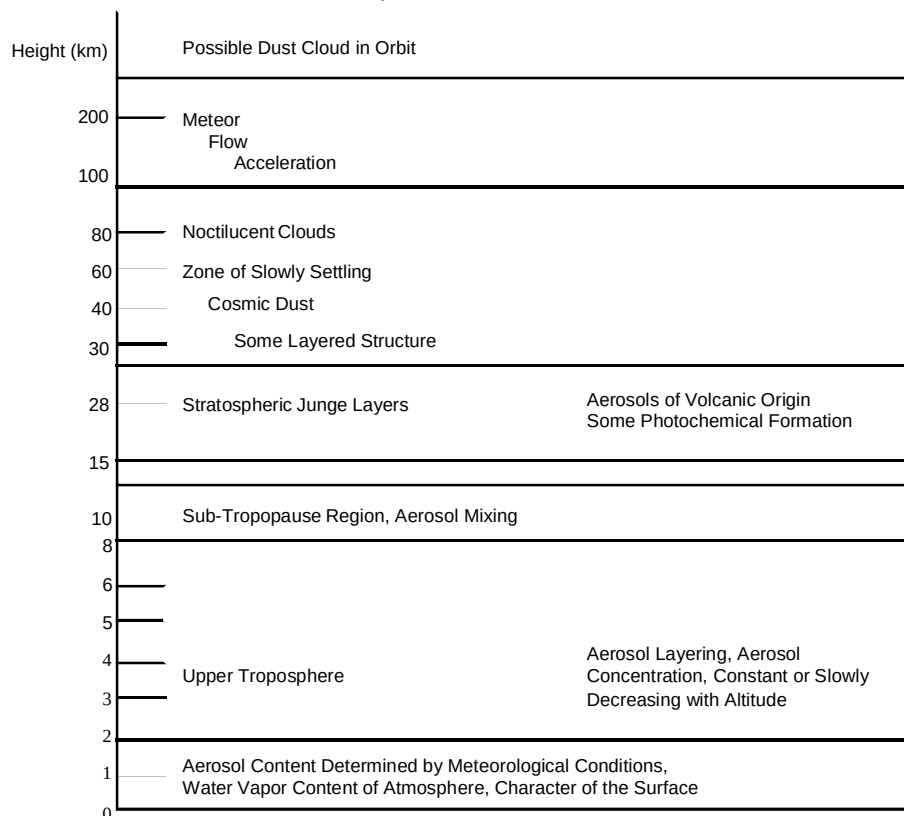


Figure 1. Characteristics of the atmospheric aerosols as a function of altitude. [Modified from *Shettle, 1989*.]

2.2 Aerosol Size Distribution

2.2.1 Characterization of Atmospheric Aerosols

Aerosols are characterized by their physical and chemical properties, namely, size, shape, and material composition. The properties of atmospheric aerosols not only vary within a unit volume of the atmosphere, but also vary considerably over the global atmosphere. Of these, traditionally the most commonly used aerosol property is the particle size. It is assumed that the shape of atmospheric aerosols is spherical, which is a valid assumption for most applications of interest, so that the size can be represented by the parameter, r , for the particle radius. (The non-sphericity of atmospheric particles becomes important in dealing with snowflakes, ice crystals, rain drops, volcanic ash, etc.)

When neither the shape, size, nor the constitution of the aerosol particles is uniform, it is referred to as *polydisperse aerosol* (a term ethnologically [Deirmendjian, 1969] derived from the classical concept of light dispersion by a prism). However, if a polydisperse suspension has particles that are uniform in physical constitution and shape but *vary in number concentration depending on the size only*, it is referred to as a “*polydispersion*,” and, if all particles have the same size or a very narrow range of sizes, then the aerosol is referred to as “*monodisperse aerosol*” or “*monodispersion*.”

2.2.2 Aerosol Size Distribution Functions

It is convenient to characterize the polydispersity of atmospheric aerosols by a distribution function, called the *size distribution*, which gives the variation of the particle size in terms of either number concentration or surface area, or volume, with size being represented by either radius (r) or logarithm (natural or to the base 10) of the radius. Before

discussing the relative merits and applications of these three representations, it is in good order to present the definition of size distribution in terms of the number concentration.

2.2.2.1 Number Size Distribution and Cumulative Number Size Distribution

The nomenclature used to represent the particle size distribution and the cumulative size distribution in the literature on the subject varies somewhat. Therefore, in order to avoid confusion, we define below the symbols, terms, and concepts that we will use and how they relate to concepts used by others to characterize these two distributions as well as altitude-size distributions.

The particle size distribution is frequently represented by [Green, A.E.S., A. Deepak, and B.J. Lipofsky, Interpretation of the Sun's Aureole Based on Atmospheric Aerosol Models, *Applied Optics*, 10 (1970), 1263–79]:

- i. The *number-radius distribution*, $n(r)$ [$m^{-3}\mu m^{-1}$], which is defined as the number of particles per unit volume within a unit radius range at r , where r is the particle radius in micrometers (μm). This quantity may be defined by the expression:

$$n(r) = dN_u(r)/dr, [m^{-3}\mu m^{-1}] \quad [1]$$

where $N_u(r)$ is the *cumulative undersize distribution*⁵. The cumulative distribution function represents the total number of particles per cubic centimeter that have radii less (greater) than r and is represented by $N_u(r)[N(r)]$ and is called the cumulative undersize (oversize)⁵ distribution function. In essence

$$N_u(r) = \int_0^r n(r)dr [m^{-3}] \quad [2]$$

$$N(r) = \int_r^\infty n(r)dr [m^{-3}] \quad [3]$$

Unless stated otherwise, the cumulative oversize distribution, $N(r)$, is considered in this work, and definitions of other quantities is based on it. Also, the number-radius distribution, $n(r)$, will be referred to as the size distribution. Note that $N(0)$ would then be the total density of the particles. Note also that

$$n(r) = -dN(r)/dr, [m^{-3}\mu m^{-1}] \quad [4]$$

so that size distribution $n(r)$ is sometimes referred to as the *differential particle number distribution*.

- ii. The *log number-radius distribution*, $N_L(r)[cm^{-3}]$, that is given by

$$N_L(r) = dN_u(r)/d(\log_{10} r), [m^{-3}] \quad [5]$$

This nomenclature was introduced by Junge, who preferred the use of the logarithmic scales because of the wide range of particle sizes and concentrations involved in representing the polydispersity of the atmospheric aerosols. It is simple to show that

$$N_L(r) = 2.306r n(r) \quad [6]$$

Various derived parameters are also frequently quoted; these include

1. the total particle number, N_o , e.g., $N(0)$ for $r = 0$ from Eq. [3]
2. the mean radius $\bar{r} = \int_0^\infty rn(r)dr / \int_0^\infty n(r)dr$
3. the effective radius $r_{eff} = \int_0^\infty r^3 n(r)dr / \int_0^\infty r^2 n(r)dr$
4. mode radii, values of r where $dN/dr = 0$
5. Particle diameter $D = 2r$.

2.2.2.2 Commonly Used Analytical Size Distribution Models

Atmospheric aerosols typically have sizes in the range 0.01–10 μm , and their number distribution across this range may be very variable. For purposes of representation and intercomparison, it is common to represent these size distributions by suitable approximating mathematical functions. Three commonly used functions are shown in Table

4. A set of eight such mathematical functions are described in *Deepak and G.P. Box [1979]*. Real distributions are often represented by a combination of one or more of these functions. The form in which these functions are presented may also differ from one author to another. In Table 4, the distributions are expressed in terms of the variation with particle radius, r , of the cumulative particle number N (i.e., the total number of particles with radii greater than r per unit volume).

Table 4. Commonly Used Size Distribution Functions (Formula Taken From *Rosen and Ivanov [1993]*)

Type	Function	Adjustable Parameters
1. Junge Power Law	$\frac{dN}{dr} = Ar^{-p}$	A, p constants
2. Log Normal	$\frac{dN}{dr} = ar^{\alpha} \exp(-br^{\gamma})$	A, constant r_g , mode radius σ_g , geometrical standard deviation
3. Modified Gamma	$\frac{dN}{dr} = \frac{A}{r \ln(\sigma_g)} \exp \left\{ \frac{-\ln^2(r/r_g)}{2(\ln^2 \sigma_g)} \right\}$	a, b, α , γ , constants

Legend: N is the cumulative particle number distribution for particles with radius greater than r .

The simplest function in Table 4 is that due to *Junge [1952, 1958]*, which has only two adjustable parameters. The exponent p is normally positive, particle number thus increases with decreasing particle radius. Use of a single positive value of p would result in an infinite total number of particles. In practice, the law may be used in a segmented form in which different values of p are used over different radius intervals, p being zero for the smallest particle (e.g., *Gras and Michael, 1979*).

The Junge power law, in its basic form, has no maximum, except at $r = 0$. Atmospheric size distributions frequently show modal structure and may be better represented by a log normal distribution or a combination of such distributions and this is probably the distribution in most common use [*D'Almeida et al., 1991; Jaenicke, 1993*]. The distribution has three constants: A , which relates to the total particle number, r_g the mode radius, and σ_g which determines the width of the size distribution. The third distribution shown is the modified gamma distribution proposed by *Deirmendjian [1963, 1964]*. This has four adjustable constants and is frequently applied to particle size distributions in haze and clouds [*McCartney, 1976*].

2.2.2.3 Surface and Volume Size Distributions

Each of the three representations of aerosol size is convenient or suitable for understanding and/or describing different aerosol properties or interactions. It is not always convenient nor suitable to present aerosol size information in terms of the number distribution. For applications to chemical interaction, surface area is more suitable, and for the discussion of mass loading and conservation, the volume distribution is the most suitable. Optical properties relate more closely to both the area and volume distributions than to the number distribution. Figure 2 shows the number, surface, and volume distributions for a typical urban aerosol distribution [*Whitby, 1975*]. The number distribution is clearly uni-modal with a peak near to a particle diameter of $0.01 \mu\text{m}$. The distribution is not symmetric but has a tail extending to larger particle sizes. As we proceed to the area and volume distribution, new modes emerge from this tail, which are not evident in the number distribution.

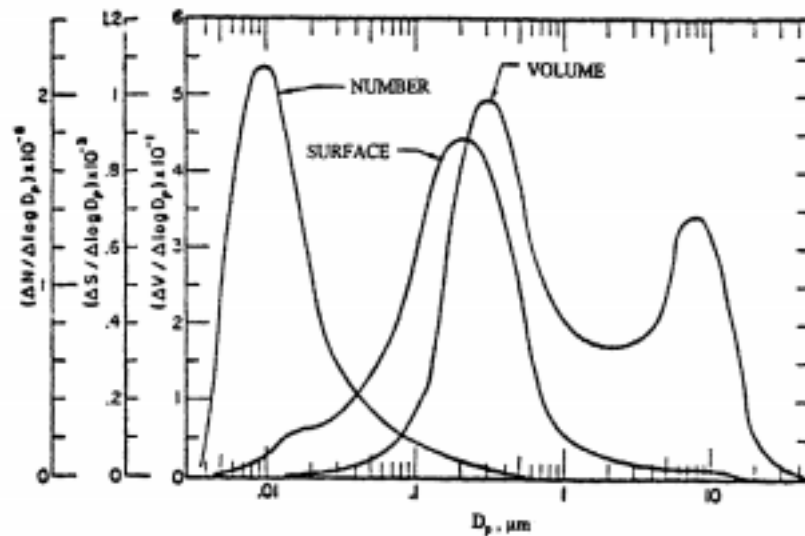


Figure 2. Normalized frequency plots of the number, surface, and volume distributions as a function of particle size for the grand average 1969 Pasadena smog aerosol. (From Prospero et al., 1983; Whitby, 1975.)

Real aerosol size distributions frequently show a trimodal structure as evidenced in Figure 2 [Prospero et al., 1983; Johnson, 1993]. This is also shown schematically in Figure 3 which depicts the principal mechanisms responsible for each of these modes. The mode with the smallest radius, namely, the nucleation mode (also referred to as Aitken or transient nuclei), represents particles that are formed by condensation and coagulation from the gaseous phase. Particles in this mode tend to grow by coagulation to form large particles in the so-called accumulation mode. The largest group of particles shown in Figure 3 (coarse particle mode) are directly produced from the land surface, the ocean, and as volcanic ash. Figure 3 also shows the primary removal mechanisms for particles in these modes. Those in the nucleation mode evolve into the accumulation mode, where (in the troposphere) they are removed by cloud and rain processes. Larger particles tend to disappear by sedimentation, as do accumulation mode particles in the stratosphere.

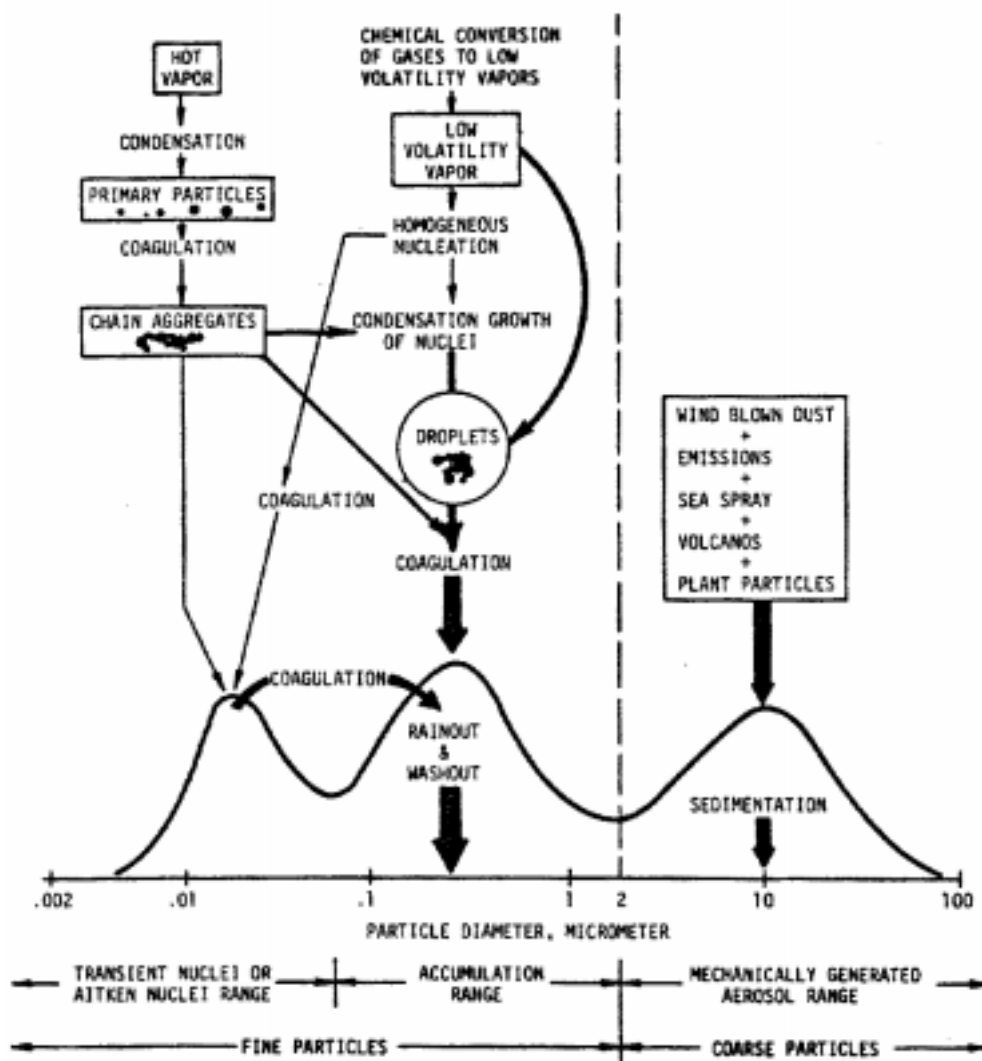


Figure 3. Atmospheric aerosol surface area distribution showing the principal modes, the main source of mass in each mode, and the principal removal mechanisms [From Prospero et al., 1983; Whitby and Cantrell, 1976].

2.3 Aerosol Types

It is conventional to divide aerosols into a number of types dependent upon the regions of the earth's surface or atmosphere in which they are to be found. The number of types and sub-types may vary somewhat between authors, but the main categories have general acceptance. Table 5 shows a list of such aerosol types generated by Jaenicke [1993] for the tropopause, to which has been added four more types for the upper atmosphere. This table also contains a brief summary of the regions of the earth or atmosphere in which these types most commonly occur and the conditions under which they are formed.

Table 5. Aerosol Types (Adapted from *Jaenicke, 1993*)

Type	Characteristics
A. TROPOSPHERIC	
Polar	Aerosols close to the surface in the Arctic and Antarctic regions. Aerosols are well-aged. Hemispheric differences exist.
Background	Typical of mid-tropospheric conditions, seen near surface is subsiding air masses or at mountain sites.
Maritime	Found at lower altitudes over the oceans. In remote locations (e.g., southern hemispheric oceans) composed of background aerosol plus sea salt. May also contain mineral dust and continental pollution in other regions.
Remote Continental	Aerosols close to surface in areas away from human development, e.g., that near forest. May contain biogenic material.
Desert Dust	Naturally occurring aerosols, wind derived from desert surfaces. Often have high mass concentrations depends upon wind speed.
Rural	Continental aerosols with moderate anthropogenic impact.
Urban	Dominated by anthropogenic emissions from industry, homes, and traffic.
B. STRATOSPHERIC	
Background	Aerosols found under non-volcanic conditions. Data from 1979 frequently cited as an example of background conditions.
Volcanic	Aerosols found in the stratosphere when perturbed by fresh volcanic injection. Consists of larger sulfuric acid droplet and ash (for very recent eruptions).
Polar Stratospheric Clouds	Found in winter at high latitudes, composed of solid nitric acid and ice.
C. MESOSPHERIC	
Noctilucent Clouds	Ice particle at high latitude summer mesopause.

Seven tropospheric aerosol types are shown in Table 5. Four of these (Polar, Maritime, Remote Continental, Desert Dust) relate to the main divisions of the earth's surface where there is little or no anthropogenic influence. Northern hemisphere Polar does not fit well into the category as there may be considerable transport of pollution into this region. Two (Urban and Rural) have respectively, major and minor anthropogenic components, and one (Background) relates to the mid-troposphere and is usually seen at the surface only at mountain locations. Two stratospheric types are shown respectively describing the stratosphere under conditions of volcanic perturbation by direct injection and under quiescent background conditions. A third stratospheric type describes polar stratospheric clouds (PSCs), which are formed in the wintertime Arctic and Antarctic regions at very low temperatures. The final type is for much higher altitude clouds forming near the mesopause and seen from the earth's surface as noctilucent clouds.

The classifications just discussed are formed in terms of global aerosol distributions and sources and, to some extent, they also reflect aerosol composition. Nevertheless, most aerosols are mixtures of components with different chemical compositions and it is necessary also to classify these aerosol components. Such a list, derived in somewhat simplified form from a list given by *D'Almeida et al. [1991]*, is shown in Table 6(a). While a few of them, such as soot and sulfuric acid, reflect fairly basic chemical compositions, most of these components consist of very complex and somewhat variable chemical mixtures which, nevertheless, have been identified as major fractions of the common aerosol types. Despite the variable nature of these components, average physical and optical characteristics have been measured and commonly are used in the modeling of aerosols. Tables of these characteristics will be presented later. Table 6(b) shows the components which make up each aerosol type listed in Table 5. Where possible, approximate component amounts have been specified and two of the aerosol types (Polar

and Maritime) have been further subdivided to reflect different amounts of aerosol transported from outside into the regions where the aerosol types exist.

Table 6. Aerosol Components and Contribution to Aerosol Types [From *D'Almeida et al., 1991; Jaenicke, 1993*]

(a) Aerosol Components		
1. Dust-like		
2. Water-soluble		
3. Soot		
4. Sea-salt		
5. Mineral		
6. Sulfuric Acid		
7. Volcanic ash		
8. Meteoric		
9. Sulfate		
10. Biogenic		
(b) Component Structure of Aerosol Types shown in Table 5		
<i>A. Tropospheric</i>		
Polar	–Antarctic	.99 (6) + 0.005 (4) + 0.005 (5)
	– Arctic	var. (3) + var. (4) + var. (5) + var. (9) [var. $\equiv$ variable]
Background		var. (6) + var. (9)
Maritime	-Clean	0.6 (4) + 0.4 (9)
	– Mineral	var. (4) + var. (5) + var. (9)
	– Polluted	0.6 (2) + 0.4 (3) + trace (4)
Remote Continental		var. (1) + var. (2) + var. (10)
Desert Dust		(5)
Rural		0.94 (2) + 0.06 (3) + trace (1)
Urban		0.6 (2) + 0.4 (3) + trace (1)
<i>B. Stratospheric</i>		
Background		(6)
Volcanic		var. (6) + var. (7)
Polar Stratospheric Clouds		var. Nitric Acid Trihydrate + var. Ice + trace (6)
<i>C. Mesospheric</i>		
var. (8) + Ice		

As the names of the various aerosol types imply, they are often confined to specific regions of the globe. Actual distributions of specific aerosol types are difficult to measure on a global scale although there may be much regional documentation. In order to estimate the global distribution, models incorporating source strengths, transport and loss mechanisms and, where appropriate, chemical transformation, nucleation, and coagulation have been developed. The predictions of one such model for desert aerosols is shown in Figure 4. This figure (after *Jaenicke, 1993*) is based on a model described by *Zimmermann et al. [1989]*. This distribution was obtained [*Wefers, 1990*] by adapting a three-dimensional climatological transport model for the desert aerosols. The aerosols in that model are followed in 10 size ranges between 0.1 and 100 μm radius. The activities of the sources are calculated from wind velocities. The sinks include only the settling velocity of the aerosols. Thus, the model is more realistic in dry than in wet geographical regions. The highly non-homogeneous distribution is very evident, major

concentrations occur over the deserts themselves, but there is evidence (supported by direct measurements) for long-distance transport over the oceans. Similar distributions are shown by *Tegen and Fung [1994]*, where the aerosols have been classified by size. Similar regional scale models have been developed for the more complex case of pollution derived aerosols. It is important to note that long-range distributions are strongly dependent upon not only the sources and wind direction, but also upon the atmospheric removal processes, particularly those associated with clouds and precipitation.



Figure 4. Mass concentration of mineral aerosols (in $\mu\text{m m}^{-3}$) for April at the 1000 hPa pressure level [Wefers and Jaenicke, 1990].

2.4 Aerosol Variations

The concentration and distribution of aerosols in the atmosphere is extremely variable. Even after aerosol formation has taken place, further modification can occur, particularly, as a result of cloud processes which are also responsible for much of the removal of aerosol from the atmosphere. Figure 5, taken from *Prospero et al. [1983]*, depicts the atmospheric processes of aerosol modification and removal. This figure emphasizes the role of clouds in aerosol conversion and removal processes. Box 5 is subdivided into 5a and 5b to denote those aerosol species that are effective as cloud condensation and ice nuclei. Note that super-cooled droplets (6a) will be converted to ice crystals (7) if they make contact with a freezing nucleus (5b). The surface area of cloud droplets (6) and ice crystals (7) in a typical cloud is tremendous; consequently, the sorptive removal of gases (1 and 2) is facilitated. Because most clouds dissipate through evaporation (i.e., the reverse of arrows 5a-6 and 5b-7), a modified aerosol population will be injected back into the atmospheric aerosol system (box 5). Rain (8) and snow (9) can also evaporate in the atmosphere, but this process is not nearly as common as cloud evaporation. Aerosols can also be deposited directly to the earth's surface by diffusion, impaction, and sedimentation; however, precipitation is responsible for the removal of about 80-90% of the submicron mass from the atmosphere. Some dry removal of aerosol occurs but the bulk of removal is by precipitation. *Jaenicke [1993]* estimates that aerosol pass through clouds about five times before precipitation occurs. This absorption and re-evaporation may produce considerable change in the aerosol physical characteristics. Jaenicke estimates that 3000 Tg per year of aerosol is "produced" from evaporating clouds, a figure comparable to those shown for other sources in Table 3.

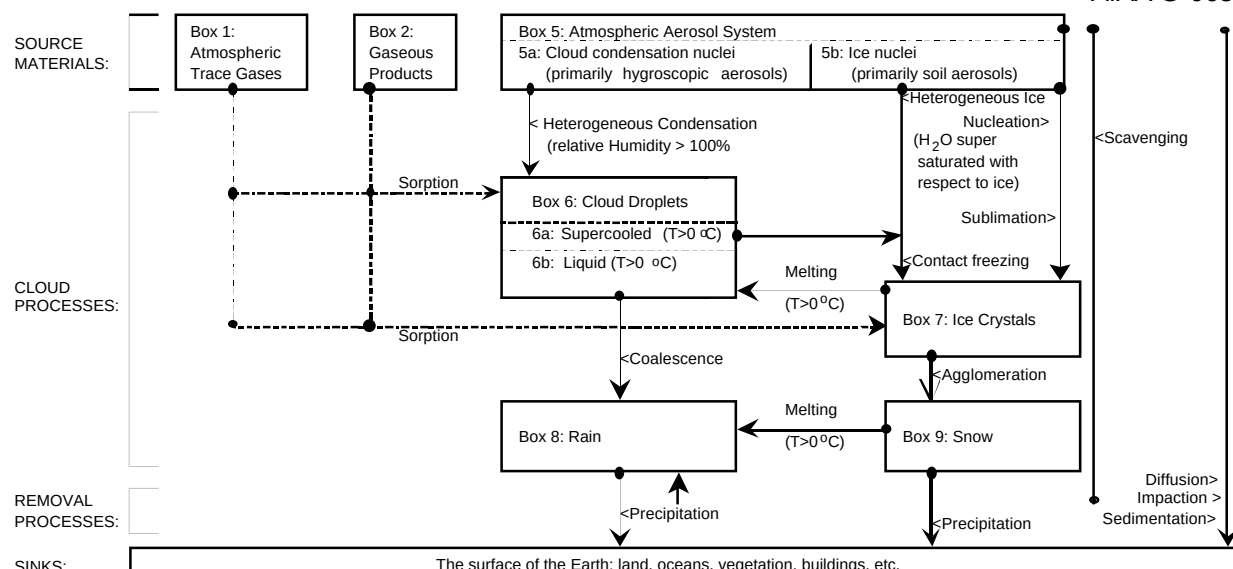


Figure 5. Aerosol modification and removal processes. [From Prospero et al., 1983.]

Residence times for aerosols in the atmosphere is dependent upon their size and altitude. This is shown by Figure 6 (after Prospero et al., 1983). The figure also shows recent estimates of the residence times of specific classes of aerosol particles (hatching [Hofmann and Rosen, 1981] and arrow [Pruppacher and Klett, 1978]). The larger particles disappear by sedimentation, the smaller particles by coagulation, resulting in the formation of longer-lived particles of intermediate size. Jaenicke [1988] has proposed the formula

$$\frac{1}{\tau} = \frac{1}{K} \left(\frac{r}{R} \right) + \frac{1}{K} \left(\frac{r}{R} \right)^{-2} + \frac{1}{\tau_{\text{wet}}} \quad [7]$$

to describe the mean residence time, τ . In this expression, K is a constant ($1.28 \times 10^8 \text{ s}$); R is the radius of the particles with the maximum residence time ($R = 0.3 \mu\text{m}$); and τ_{wet} is a measure of the influence of wet removal ($\tau_{\text{wet}} = 8 \text{ days}$, in the lower troposphere; and, $\tau_{\text{wet}} = 3 \text{ weeks}$, in the middle to upper troposphere). This yields a maximum residence time in the lower troposphere of about a week, increasing to several weeks in the upper troposphere. In the stratosphere, residence times are typically of the order of 12 months for volcanic aerosols.

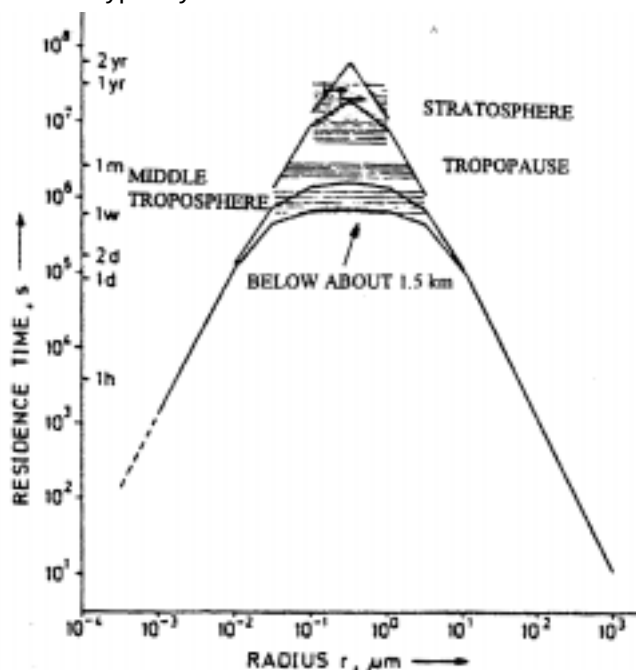


Figure 6. Aerosol residence time of individual particles as a function of particle size and altitude in the atmosphere [Prospero et al., 1983; Jaenicke, 1980].

Water soluble aerosols change their radius in response to changes in atmospheric humidity. *Hänel and Bullrich [1978]* and *Hänel and Lehmann [1981]* discuss these variations and present compilations of radii versus relative humidity for different aerosol compositions. A rough approximation for the equilibrium radius versus relative humidity can be obtained using:

$$\ln f = \frac{(A/r_o)}{(r/r_o)} - \frac{B}{(r/r_o)^3 - 1} \quad [8]$$

f = relative humidity as pressure ratio (actual to value when saturated)

r = equilibrium radius of the wet particle

r_o = radius of the dry particle

A and B are constants

Values for f and r/r_o are tabulated for four different classes of aerosols by Hänel and Bullrich (1978). An example of one of the tables for continental background aerosol is reproduced as Table 7. Note the difference in values when f is decreasing and increasing and the small radius change for values of less than about 0.6.

Aerosol concentrations also change in response to episodic events. These include desert storms, fires, and volcanic eruptions. Desert dust storms and fires related to burning for agricultural purposes show regular seasonal cycles, and statistical predictions can be made about their likely impact on the troposphere. Volcanic eruptions are random in magnitude and time and, as a result, stratospheric aerosol concentrations are unpredictable, even though, under background conditions, there are regular seasonal changes. Other episodic events, such as naturally occurring fires (forest, oil, etc.), industrial catastrophes, etc., all produce nonpredictable changes in aerosol concentrations. Climate changes, either natural or associated with human activity, can also be expected to cause long-term regional changes in aerosol amounts.