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Coastal Aerosol Workshop Proceedings 23-24 May 1994

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The Coastal Aerosol Workshop (23-24 May 94, Monterey CA) was held to assess the state of the art of aerosol research, identify deficiencies in current knowledge of coastal aerosols, and define fruitful areas for a Navy research program. Workshop results will form the basis for a coastal aerosol research program conducted jointly by the Naval Research Laboratory and Office of Naval Research. Workshop sessions considered three component topics: aerosol surface source functions; aerosol physics, chemistry, and transformation processes; and aerosol radiative properties. Issues identified as having pressing requirements for further research included: (1) Surface aerosol generation - spume generation, surf and near-shore processes; (2) Transformation processes - near-surface processes' dynamics and characterization; (3) Remote sensing - programs integrating aerosol, gas, and meteorological measurement suites with satellite- and ground-based remote sensing; (4) Modeling and meteorology - initialization of aerosol distributions; and (5) Aerosol technology - need for a utilitarian LIDAR system capable of multi-parameter inversion.

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COASTAL AEROSOL WORKSHOP PROCEEDINGS

Executive Summary

The purpose of the Coastal Aerosol Workshop was to assess the state of the art of aerosol research, to identify deficiencies in current knowledge of coastal aerosols, and to define fruitful areas for a Navy research program. The results of the workshop will form the basis for a coastal aerosol research program to be jointly conducted by the Naval Research Laboratory and the Office of Naval Research; the former will conduct primarily in-house research, and the latter will sponsor academic and industrial research.

The workshop participants included scientists currently active in areas related to coastal aerosol research. They represented a cross-section of Navy scientists and managers, and researchers from academic and industrial laboratories. The workshop consisted of plenary sessions designed to provide an overview of Navy plans and operational needs that drive the requirements for basic research in coastal aerosol processes. These were followed by general reviews of research programs in marine aerosols, and applications of this research to operational models. The participants then met in working sessions as three separate groups - aerosol surface source functions; aerosol physics, chemistry, and transformation processes; and aerosol radiative properties - after which group representatives presented findings in a plenary session.

Following the group reports, the integration of the Navy Operational Regional Atmospheric Prediction System (NORAPS) was described. This integration involves the incorporation of the various interdisciplinary processes into a numerical meteorological aerosol

model, and a detailed analysis of performance over a variety of conditions and domains. The findings of the groups were then reviewed in the context of developing a capability for diagnosing and forecasting aerosol concentrations and properties in coastal regions.

The conclusions of the groups' discussions were separated into five topical areas with groups identifying the issues considered most important within each area. This prioritization was based on the criteria that the research area is important in the understanding of coastal aerosol characteristics and processes, and that this area is underemphasized in current research programs. The issues identified as having the most pressing requirements for further research:

- Surface aerosol generation: spume generation; surf and near shore processes.
- Aerosol transformation processes: dynamics and characterization of near surface aerosol transformation processes
- Remote sensing: need for closure experiments; programs must integrate aerosol, gas, and meteorological measurement suites with remote sensing (satellite and ground based),
- Modeling and meteorology: initialization of aerosol distributions
- Aerosol technology: need for utilitarian LIDAR system capable of multi-parameter inversion

1. Background

The Navy must be capable to diagnose and predict the impact of aerosols on systems operating in visible and infrared (IR) wavelengths, and improve system design and performance based on a more accurate knowledge of the environment. The Navy has responded to these needs with a continuing investment in aerosol research, which has led to the development of an empirical operational model that predicts the vertical distribution of aerosol size spectra, from which electro-optical extinction is calculated. Because this model is one-dimensional, it performs best under open ocean conditions where ocean surface-source aerosols predominate and weather conditions are to a great extent horizontally homogeneous. The model performs poorly in coastal regions where continental and anthropogenic aerosols complicate the aerosol source distribution functions, and conditions are often anisotropic and nonstationary. Recent advances in aerosol science have progressed to the point where it is possible to start the design of a process-oriented dynamic aerosol model coupled with a high resolution meteorological/oceanographic model. Such a model would improve the existing empirical model by enabling a wider applicability to more diverse topographic and environmental conditions, as well as providing a true forecast capability.

As a result of the Navy's shift of investment strategy towards the coastal domain, the Naval Research Laboratory (NRL), and the Office of Naval Research (ONR) are jointly developing a plan for a new aerosol initiative designed to improve aerosol diagnosis and prediction in coastal regions. An important part of the new initiative would be the development of a coastal aerosol model (CAM), which would use available surface and remote observations with environmental and aerosol process models to predict aerosol concentration in the

atmospheric boundary layer. The architecture supporting the CAM will require a keen understanding of the maritime sources of aerosols as well as the source transformations, chemistry, and disposal processes associated with continental aerosol advected from land to sea. Ultimately the CAM will require development of high resolution models, initialization schemes (requiring a variety of remote and local measurements), and field validation experiments using in situ and remote measurements.

A workshop was conducted in Monterey, California on May 23-24, 1994, to outline research requirements for the development of a coastal aerosol model. The specific goals of this workshop were to assess the state of the art of aerosol research, to identify deficiencies in current knowledge of coastal aerosols, and to define fruitful areas for a Navy research program. Invitations to participate were primarily sent to NRL and ONR contract scientists conducting research on aerosol processes and related topics. The workshop represented a cross-section of Navy scientists and managers together with researchers from academic and industrial laboratories. The workshop announcement is included as Appendix A. Workshop participants are listed in Appendix B.

The workshop agenda is shown in Appendix C. Initial discussions were designed to provide a baseline review of Navy needs and current aerosol research. Copies of the transparencies presented in these reviews are included in Appendix D. The formal presentations were followed by separate topical meetings to determine deficiencies and provide research recommendations in more specific aerosol research areas. The conclusions of the working groups were presented in a series of transparencies included as Appendix E. The participants met again in a plenary session to review the separate topical meeting findings and to hear a presentation

describing NRL combined aerosol and meteorological modeling. This modeling discussion served to integrate the separate discussions of meteorology, chemistry, physics, optics, and oceanography toward the goal of environmental forecasting and analysis. The participants' final action was to review the separate topical recommendations in the context of program goals and combine and refine these to form a final set of recommendations.

2. Review Session

The Navy is planning a rapid increase in the deployment of tactical systems operating with short wavelength (optical) active and passive sensors. With the changing global political considerations, these systems are increasingly operating in coastal regions. Current environmental effect predictions are mainly designed for open ocean regions and are not applicable to the more complex coastal zone. Thus the Electro-Optical Tactical Decision Aid (EOTDA), for example, is not particularly successful in predicting the environmental degradation to system performance in coastal regions. This deficiency is being addressed by an increased emphasis on research to develop an effective basic understanding of the physics and chemistry of coastal aerosols, and the expected development of a 3-D prediction capability based on research results.

The aerosol review session described current understanding of maritime surface marine aerosols, with following presentations reviewing the status of process oriented aerosol modeling and the supporting measurement programs. The session concluded with a description of the operational aerosol concentration and extinction algorithm (Navy Operational Vertical Aerosol Model, or NOVAM). The transparencies from the presentations are included as Appendix D.

3. Working Sessions

Participants then separated into three working groups each of which was separately tasked to review the state of the art and identify areas that require additional research. The groups returned with presentations summarizing their discussions. The working groups' reviews

of current research deficiencies and recommendations for resolving these deficiencies are discussed in the remainder of this section.

3.1 Aerosol surface source functions

The aerosol source function is vital to an understanding of the behavior of the coastal atmospheric aerosol distribution. The current knowledge of marine aerosol sources is based on observations and modeling of the source function over the open ocean. Aerosol sources on land which contribute to the marine aerosol distribution have been inadequately addressed, except for a limited number of studies conducted in the region of the Los Angeles basin. The limitations of the current state of the art knowledge of aerosols in the coastal zone (within 100 km of the coast, both landward and seaward) were summarized to be in several phenomenological areas. Recommendations were then provided in each of these areas.

3.1.1 Limitations of current knowledge.

Bubble generated aerosols

The understanding of bubble mechanisms for aerosol generation has improved substantially over the last 20 years. The source function for jet drops is now known to within an order of magnitude. Due to technological limitations, film drops are not as well known. We can expect the modeling to attain closure within 5 to 10 years , providing a reasonably consistent understanding of this aerosol generation process. This could be accomplished with the development of improved technology and well formulated experiments.

Spume is created when high winds physically tear and remove the wave crest, resulting in the generation of large numbers of aerosol particles. The source function is recognized as important, but is not known to within 3 orders of magnitude. Based on energy arguments, the

source function has an assumed wind dependence expressed as approximately the third power of the friction velocity, u_* . The understanding of these processes is about a decade behind that of bubble generated aerosols.

Surf zone generation

Surf zone aerosol generation processes are poorly understood. The expectation is that wind, wave height, shoreline slope and thermal stability have significant effects, but none of these dependencies is quantified. Initial estimates suggest the surf zone could be a significant source of coastal zone aerosols. It is hypothesized that the surf zone processes strongly influence characteristics of the coastal zone aerosols in moderate and light winds, although this hypothesis has not been tested.

Experimental measurements

Simultaneous water bubble populations and atmospheric aerosol population measurements are required both in the laboratory and in the field. These are needed to improve our understanding of the physics of bubble mediated aerosol production under open ocean and coastal conditions

Modification by surface waves

The near surface aerosol distribution may be significantly modified by the wave field. The wave field is likely to play a significant role in modifying the bubble mediated aerosol source function. The air flow in the wave region controls aerosol motion, and as such has a strong impact on transport and deposition of the aerosol. These processes have not been studied or quantified.

Contaminants

Observations and theoretical work indicate that surface contaminants may have a significant effect on the surface wave field, the air-sea exchange processes, and the droplet surface tension. The reduced air-sea transfer of gas species is likely to affect the aerosol distribution as well as the size dependent composition. Surface contaminants and high concentrations of bulk organics are prevalent in coastal waters, and would probably profoundly modify sea surface aerosol production.

Rain

Rain induced droplets, via splash or intermediate transfer, may be episodically important. Washout and rainout processes would strongly change both the size distribution and composition of the base aerosol distribution. This could be especially important in coastal regions where orogenic precipitation is frequent.

Spatial variability

The current aerosol models are based on a uniform open-ocean model of the atmosphere. In coastal regions, the atmospheric conditions will be much more variable because of complex coastline topography, sea breezes, clouds, and/or local meteorological phenomena. In addition, coastal aerosol sources may result in extreme changes in aerosol distributions over fairly small distances.

3.1.2 Recommendations

This working group concluded that the following efforts were important for the understanding of coastal aerosol processes:

Surf aerosol sources

We need to study specific mechanisms which generate spray droplets ins surf and other coastal regions. Investigations should be conducted in the laboratory and in the field.

Spume aerosol source

We need to study the production of spume droplets in the maritime atmosphere. Experimental measurements should be conducted both in the field and in the laboratory, especially since controlled high wind measurements are notoriously difficult to conduct in the field. The results should elucidate the physical mechanisms governing spume droplet production.

Breaking wave aerosol source

We need a detailed investigation of the bubble and droplet fluxes in breaking waves. These studies should also be conducted both in the laboratory and in the field.

Parameters for aerosol nowcast

We must develop a finite list of the parameters that are required for an aerosol nowcast. These parameters should be directly or almost direct measurable by local or remote techniques.

3.2 Aerosol physics, chemistry, and transformation processes

This working group addressed issues regarding the physics, chemistry, and transformation processes that determine the size and composition of atmospheric aerosols in the coastal and marine environment. The issues were categorized as being related to sources, transformations, and sinks. In the two and one half hours allotted for deliberations, only the most important issues could be considered. The following discussion addresses topic areas the group agreed were important; further comments and discussion are added by the editors.

3.2.1 Limitations of current knowledge

Direct injection of aerosols

Aerosol particles directly injected into the atmosphere represent one of the major components of the coastal aerosol distribution. As such any modeling will require the size resolved physical, chemical and optical properties of aerosol generated in coastal regions. Among the sources for the aerosols are the urban plume, desert dust, biomass burning, sea surface generation, and sea surface emission of relevant precursor gases. These local sources are supplemented by advected aerosols, both from continental and maritime regions.

Nucleation

The dominant process replenishing submicron aerosols and cloud condensation nuclei in the remote ocean environment is nucleation. Nucleation is generally believed to involve gas phase conversion of SO_2 and/or dimethyl sulfide (DMS) to sulfuric acid, and is currently represented in models as binary homogeneous nucleation of sulfuric acid droplets. It is not clear that nucleation is binary, and theory suggests other mechanisms may be energetically preferred. These other mechanisms are not yet sufficiently understood to be included in models. Other possible products of nucleation products are NH_4Cl, which can be due to biogenic emission of ammonia. Furthermore, it is possible that organics play a role in nucleation, but the processes are not adequately understood.

Cloud processing

Absorption of soluble trace gases by cloud droplets and subsequent removal by precipitation is an important process for scavenging gases from the atmosphere. It has recently been demonstrated that these liquid phase reactions may be accompanied by a more subtle

process, in which some of the lower volatility reaction products are recycled back into the atmosphere as particles with increased mass. This changes the characteristics of the size distribution substantially, and thus would have a significant effect on optical extinction at visible wavelengths. The reaction rates constituting these processes are not yet known and further observation and analysis are required.

Heterogeneous chemistry

The physical and chemical processes occurring on particle surfaces are not known well. the condensation coefficients for important condensing species have not been calculated or measured. For example, the condensation coefficient on H_2SO_4 is variously cited as being from 0.04 to 1.0. In addition chemical reactions often occur on the surface of the aerosol, but these surface processes are not understood..

Aerosol removal processes

The main physical aerosol loss mechanisms are precipitation scavenging and surface deposition. Precipitation scavenging efficiency is expected to have a strong dependence on aerosol size and composition. The precise dependence is not currently known.

Similar surface deposition algorithms assume dry aerosols and do not consider the effects of the ambient turbulent humidity field. The loss of aerosols at the surface is known over a dry surface, but over a wet surface, particularly with an accompanying water wave field, the loss process is not known.

3.2.2 Recommendations

The recommendations of the group were categorized as laboratory studies, field measurements, and aerosol modeling. As a general comment, it was emphasized that this

program must involve different disciplines, and be an integrated unit with aerosol, air chemistry, and cloud physics expertise.

Laboratory studies

Determine dominant oxidation pathways and rate constants for DMS and SO_2.

Test proposed nucleation mechanisms.

Test theories and rate constants involved in cloud processing, including SO_2 oxidation and neutralization by NH_3, and determine rate coefficients for these processes.

Investigate the relative importance of proposed heterogeneous reaction schemes.

Field measurements

Characterize aerosol sources, including physical and chemical properties of aerosols associated with urban plume, desert dust, biomass burning, sea surface generation, and sea surface emission of relevant precursor gases.

Measure environmental conditions when nucleation occurs.

Conduct a Lagrangian aerosol experiment to follow an air mass advecting over a region with varied topographic, meteorological and oceanographic conditions.

Aerosol Modeling

The improved knowledge of the physics and chemistry of atmospheric aerosols resulting from laboratory studies, field measurements, and related theoretical studies must be incorporated into aerosol models. Process-oriented models are necessary to predict the interaction among the

individual processes, and are needed as a diagnostic tool to interpret field data and test our understanding of the processes.

3.3 Aerosol radiative properties

The aerosol radiative properties group discussed the issues involved in understanding radiative transfer through aerosol distributions. These issues were discussed in the context of the need for remote sensing of the distribution, and the requirements for understanding radiative processes in numerical weather forecast models and numerical climate change models. A number of deficiencies in the current state of the art were identified by the group.

3.3.1 Limitations of current knowledge

Long term measurement record.

There is no current long term record of aerosol properties with supporting meteorology which can be related to remote sensing. These properties include size distribution, optical depth, and single scatter albedo. A long term data base of satellite measurements is required to generate size-specific source functions in the coastal regions

Source region characterization

Aerosol source functions in the coastal regime will be a complex mixture of anthropogenic, continental, and maritime components. These models are available for only a few specific regions, and in general are not known in coastal regions. The general physics of coastal mixing processes must be determined in order to characterize the wide variety of coastal regions without a correspondingly large data base. However, this process modeling must be based on a limited coastal aerosol data base, thus making the data base extensible to more regions.

Spatial resolution requirement

Current satellite sensor resolution (e.g., 1.1 km for NOAA AVHRR, 12 km for GOES VISSR) is inadequate for coastal applications. Some higher resolution sensors are available with SPOT and LANDSAT sensors, but these do not have appropriate spectral characteristics for aerosol inversion. The NASA EOS program will have appropriate resolution and wavelength, but preparations for their utilization have only just started with the AVERS and HYDICE programs.

Aerosol models

Aerosol models are currently inadequate to describe the range of aerosol conditions observed within the coastal environment. The modeling of the composition, size, and shape as a function of conventional meteorological parameters, as well as more specific variables (e.g., chemical composition), must be improved to provide better treatment of radiative transfer in the coastal zone.

Characterization of water leaving radiance

The lack of quantitative knowledge of the water leaving radiance (WLR)is one of the major limitations to the calculation of accurate aerosol retrievals from satellite measurements. While current models assume the WLR to be constant, it is known to be a function of wind speed, sea state, wave slope, contaminants, and sub-surface conditions. In order to improve the retrieval of aerosol concentrations from remote sensing measurements, the physical relationship of the WLR to appropriate environmental parameters must be developed and tested to identify uncertainties, limitations, and improvements.

Mie scattering for non spherical aerosols

Scattering and absorption of electromagnetic waves by aerosols is usually treated using Mie theory, which assumes a uniform spherical scattering dielectric. Aerosols are often not spherical, nor uniform in their distribution of matter within the droplet. The effect of this approximation must be determined to relate aerosol extinction calculations to real world conditions.

Multiple scattering

Under most circumstances, single scattering calculations are sufficient for satellite near nadir retrievals and calculations. In the same way, single scattering is sufficient for many atmospheric transmission calculations. However, under conditions of high aerosol number concentrations or near grazing propagation paths, the effects of multiple scattering must be included.

Inversion techniques

The aerosol retrieval from satellite and LIDAR measurements depends on an inversion of the effects of scattering and absorption over the total path. The inversion techniques in current use are not always stable, so alternate, more robust methods must be determined. This may be done by specification of different spectral characteristics of the measurements, as well as by development of advanced mathematical techniques.

Closure measurements

The essential characteristic of remote sensor measurements is that each sensor has its own strengths and weaknesses. For example, one sensor may be limited in horizontal resolution but does provide excellent spectral resolution. Another sensor operates in an entirely different region

of the electromagnetic spectrum but provides a vital part of the information required for the aerosol size distribution description. The output of these sensors must be integrated, possibly using models to account for spatial, temporal and spectral differences.

3.3.2 Recommendations

Hyperspectral aerosol retrievals

The current monochromatic and bispectral aerosol retrieval should be extended to hyperspectral techniques, using an increased number of wavelengths. This has the potential for retrieving needed detail of the aerosol size distribution and aerosol composition.

Multiple scattering

Single scattering models must be extended to multiple scattering models for description of optical propagation through a field of aerosols. The requirements for use of multiple scattering models rather than the simpler single scattering models must be specified particularly for coastal conditions. In addition reliable, robust, and fast mathematical techniques are not available, and must be developed.

Water leaving radiance

The parametrization of the water leaving radiance must be developed. This includes the determination of which parameters are important, the sensitivity of the results to the use of these parameters, and the accuracy of the required parameters.

LIDAR for real time analysis

The only quantitative measurement technique for observing the four-dimensional characteristics of the coastal aerosol distribution is the LIDAR. It has been shown to be useable

in coastal regions, to provide a time-varying three-dimensional description of the aerosol distribution. The LIDAR work must be supported and improved using new and improved source and receiver technology. New algorithms for tropospheric and near-source retrievals.

Closure experiments

Field programs must be completed for the various parameters that are known to determine the characteristics of the aerosol distribution. These field programs must be designed in such a way that they form a complete set of conventional meteorology, specific aerosol physics and chemistry, and remote sensing measurements. Since experiments on this scale are probably budget-limited, different agencies must have cooperative programs including all required components.

Ground based remote sensing of model input

Numerical modeling of the aerosol concentration will probably require subsidiary input information supporting satellite measurements. This includes sea state, whitecap coverage, and presence and concentration of tropospheric molecular species, including O_3 and SO_2.

4. Concluding recommendations

The separate working group recommendations were reviewed in a plenary session of all the workshop participants. It was decided the group recommendations would be separated into five topical areas characterizing the main research requirements. This prioritization was based on the criteria that the research area was important in the understanding of coastal aerosol characteristics and processes, that the area was underemphasized in current research programs,

and that potential improvement in future Navy capabilities could be anticipated. The recommended issues with the most pressing requirements for further research were as follows:

- Surface aerosol generation: Spume generation; surf and near shore processes.

Generation of aerosols by spume is newly recognized as an important source of aerosols, and requires investigation. Surf generation of aerosols may play an important part in the description of the aerosol distribution in the entire coastal region. Process and characterization studies must be conducted to evaluate these aerosols.

- Aerosol transformation processes: Dynamics and characterization of near surface aerosol transformation processes

The coastal aerosol transformation processes are inherently more complex than those over the ocean. These processes must be closely linked to the meteorology of the region.

- Remote sensing: Need for closure experiments, integrating complete measurement suites, remote sensing (satellite and ground based) and in situ sensing

Experiments for validation of remote sensing retrievals must be complete, including all relevant meteorological parameters.

- Modeling and meteorology: Initialization of aerosol distributions

The space and time scales of the aerosol distributions must be understood to implement into a numerical model. The initialization may include the more general issue of boundary conditions which are time-dependent and anisotropic.

- Aerosol technology: Need for smaller, cheaper, and more robust LIDAR systems.

The main reason for the limited deployment of LIDAR systems is the expense and fragile nature of the instrument. Research should be conducted to determine other alternatives which can lead to durable and affordable systems.

Appendix A

Coastal Aerosol Research

Announcement of a Planning Workshop

The Navy needs a capability to assess and predict the impact of aerosols on DoD systems operating at visible and IR wavelengths. In response to this need, there has been a long tradition of Navy investment in aerosol research, and the results have led to an operational empirical model which predicts the vertical distribution of aerosol size spectra and resulting electro-optical extinction. . This model performs best under open ocean conditions where surface generated sea-salt aerosols dominate. It functions poorly in coastal regions where continental and anthropogenic aerosols predominate. Recent advances in aerosol science have progressed to the point where it is possible to think in terms of a process-oriented, dynamic aerosol model coupled with a high resolution meteorological model to supplement the existing empirical model.

As a result of the Navy's shift of investment strategy towards the littoral domain, ONR and NRL are considering a new aerosol initiative designed to improve aerosol and extinction predictions in the coastal zone. An important element of the new initiative would be the development of a coastal aerosol model (CAM). The model would use available observations (surface and remote) to analyze and predict aerosol concentrations in the atmospheric boundary layer. The architecture supporting the CAM will require a keen understanding of the marine source of aerosols as well as the source, transformation, chemistry, and sink processes associated with continental aerosol being advected from land to sea. Ultimately the CAM will require development of high resolution models, initialization schemes (possibly requiring remote sensing techniques), and field validation experiments using in-situ and remote measurements.

In order to develop the details of the architecture required to develop a first order working CAM, we must first identify and document the current research and state of the art. This information is intended to identify which components of the CAM are well understood, which are poorly understood, and which comprise new frontiers of research. The participating scientists are expected to be aware of related efforts in the broad aerosol research community.

To initiate this effort, ONR and NRL will host a two day workshop, currently scheduled for 23-24 May 1994, to be held at the Monterey campus of the Naval Research Laboratory. The workshop will co-chaired by Andy Goroch of NRL (tel. (408)-656-4889, FAX (408)-656-4769, INTERNET "goroch@nrlmry.navy.mil", A.Goroch/OMNET) and Gary Geernaert of ONR (tel. (703)-696-2496, FAX (703)696-3390, INTERNET "geernag@onrhq.onr.navy.mil", G.Geernaert/OMNET). Invitations are extended to existing ONR grantees and NRL scientists involved in the basic and applied issues associated with coastal aerosols. Attendees will be expected to review current work, identifying research needs in the context of CAM development. Outside investigators are welcome, though their participation will require permission by the workshop co-chairs.

The purpose of the workshop will be to

(1) Review and assess the present state of knowledge and importance of various physical mechanisms which determine and control the aerosol size distribution in coastal regions,

(2) Determine the feasibility of successfully developing a CAM and to determine a possible architecture,

(3) Summarize current activities,

(4) Outline research required to develop an operation CAM to different levels of accuracy,

(5) Recommend to NRL and ONR an investment strategy to develop a CAM.

PRELIMINARY WORKSHOP AGENDA

23 MAY, 1994

0800 CHECK-IN

0830 INTRODUCTION
Gary Geernaert, ONR; Andy Goroch, NRL

0910 NAVY NEEDS AND R&D REQUIREMENTS
Pete Rannelli, NRL

0940 OVERVIEW PRESENTATIONS

0940 CURRENT NAVY OPERATIONAL MODELS - Stu Gathman, NRaD

1010 COFFEE BREAK

1030 STATUS OF PROCESS ORIENTED MODELING
Jim Fitzgerald, NRL

1110 SEA SURFACE AEROSOL SOURCES
Ed Andreas, CRREL

1150 LUNCH BREAK

1300 OVERVIEW PRESENTATIONS (CONT.)

1300 AEROSOL PHYSICS/CHEMISTRY/TRANSFORMATIONS
Bill Hoppel, NRL

1330 AEROSOL REMOTE SENSING
Phil Durkee, NPS

1410 WORKING GROUPS FORM AND CONVENE
Preliminary working groups are
(A) Sea surface generation mechanisms
(B) Aerosol physics and chemistry
(C) Radiative properties and remote sensing

1700 END OF FIRST DAY

24 MAY, 1994

0800 CHECK-IN

0830 RECONVENE WORKING GROUPS

1200 LUNCH

1300 PRESENTATIONS BY WORKING GROUP (30 MIN EACH)

1430 Coffee break

1500 DISCUSSION: COASTAL AEROSOL MODEL ARCHITECTURE INCLUDING METEOROLOGICAL ISSUES

1630 SUMMARIZE RECOMMENDATIONS

1700 WORKSHOP ADJOURN

Appendix B
Participants

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Su, M. Y.	NRL SSC	(601) 688-5241	(601) 688-5997	
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Appendix C

COASTAL AEROSOL RESEARCH INITIATIVE WORKSHOP AGENDA

23 MAY, 1994

0800 CHECK-IN

0830 INTRODUCTION - Dr. Gary Geernaert, ONR; Dr. Andy Goroch, NRL

0900 NAVY NEEDS AND RESEARCH AND DEVELOPMENT REQUIREMENTS - CDR Pete Ranelli, NRL

OVERVIEW PRESENTATIONS

0940 CURRENT NAVY OPERATIONAL MODELS - Dr. Stuart Gathman, NRaD

1010 Coffee break

1030 SEA SURFACE AEROSOL SOURCES - Dr. Ed Andreas, CRREL

1110 AEROSOL PHYSICS/CHEMISTRY/TRANSFORMATIONS - Dr William Hoppel, NRL

1150 LUNCH BREAK

1300 STATUS OF PROCESS ORIENTED MODELING - Dr. Jim Fitzgerald, NRL

1340 AEROSOL REMOTE SENSING - Dr. Phil Durkee, NPS

1410 WORKING GROUPS FORM AND CONVENE

WORKING GROUPS:

(A) Sea surface generation mechanisms
Session Leader: Dr. Garrett DeLeeuw, TMI
Rapporteur: Dr. Ed Monahan, UConn

(B) Aerosol physics and chemistry
Session Leader: Dr. William Hoppel
Rapporteur:Dr. Volker Mohnen, ASRC

(C) Radiative properties and remote sensing
Session Leader: Dr. Phil Durkee, NPS
Rapporteur: Dr. Eric Shettle, NRL

1700 End of Day 1

24 MAY, 1994

0800 RECONVENE WORKING GROUPS, Review recommendations

Plenary Sessions

0830 Present and review working group recommendations

0945 Coffee break

1000 Review of aerosols in mesoscale models - Dr. Chi-Sann Liou, NRL

1040 DISCUSSION: Coastal aerosol model architecture - physical, chemical and meteorological issues.

1200 Lunch

1330 SUMMARIZE CONCLUSIONS AND RECOMMENDATIONS

1430 WORKSHOP ADJOURN

1500 Separate NRL and ONR meetings dicussing future directions and plans.

Appendix D

Transparencies of Invited Presentations

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Overview of current operational models, NAM and NOVAM.

Stuart G. Gathman
NRaD code 543

CAM planning initiative workshop

Areas covered by this overview

- History - Pre NAM models
- NAM
- NOVAM
- Limitations to NAM and NOVAM
- Areas of current thrusts
  - ANAM
  - CAM
- Recommendations

History of Aerosol applications

- Pre - Electro - Optics days
- The battle between extinction and aerosol as the mechanism for modeling
- Early aerosol models
- Navy Aerosol Model- **NAM**
- Navy Oceanic Vertical Aerosol Model **NOVAM**
- Advanced Navy Aerosol Model- **ANAM**
- Coastal Aerosol Model - **CAM**

Driving Force for Aerosol Modeling

- Electro/optical properties over the sea.
- Difficulties in transmission path measurements over the sea

Visibility and Fog

- Early issues dealing with aerosol in the fleet had to do with visibility, clouds and fog.
- Fog forecasting needed for a/c operations
- Were ships visible from a/c overhead?

Pre-NAM models

Navy Aerosol Model

- Uses available at sea aerosol measurements
- Model based on superposition of several log normal curves
- Uses precalculated Mie code to extract optical/IR data from aerosol model
- Model parameters based on observable meteorological data

Navy Aerosol Model

- How NAM is used
  - Forecast
  - Nowcast
  - Climatologies
- Insertion into LOWTRAN 6
- Availability of large size aerosol data
- Correction put into Lowtran 7

NAM/NOVAM lognormal size distribution

$$\frac{dN_i}{dr} = A_i \frac{\exp\{-1*\left[\log_{10}(\frac{r}{C_i * f})\right]^2\}}{f} \qquad i = 1,2 \text{ and } 3$$

$$A_1 = 2000 \cdot amp^2,$$

$$A_2 = MAX[5.866 \cdot (\overline{w} - 2.2), 0.5],$$

$$A_3 = 10^{(0.06 \cdot w' - 2.8)}$$

The constants C1, C2, and C3 are: 0.03, 0.24, and 2.0 respectifully.

Humidity Relationship

$$f = \left[\frac{C7 - S}{C8(1 - S)}\right]^{\frac{1}{3}}$$

S=Rel Hum (%)/100

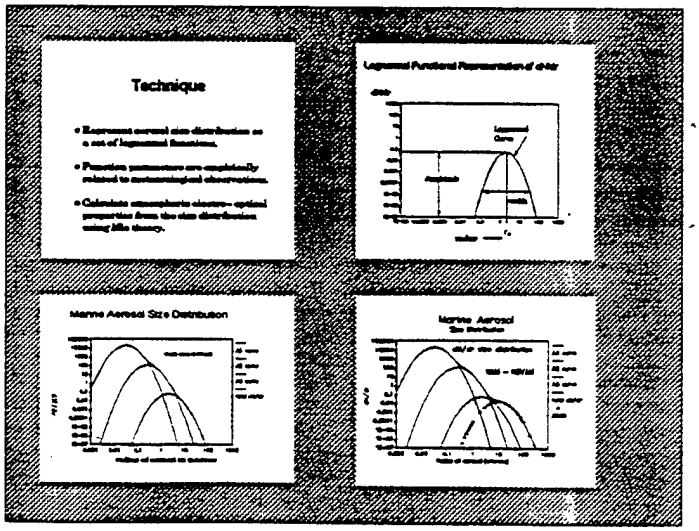
Aerosol Model	NAM Component	C7	C8	Range of Validity
Sea Salt	2	1.83	5.13	RH<99.9%
Sea Salt	3	1.97	5.83	RH<99.99%
Urban	1	1.28	2.41	RH<99%
Rural	1	1.17	1.87	RH<99%

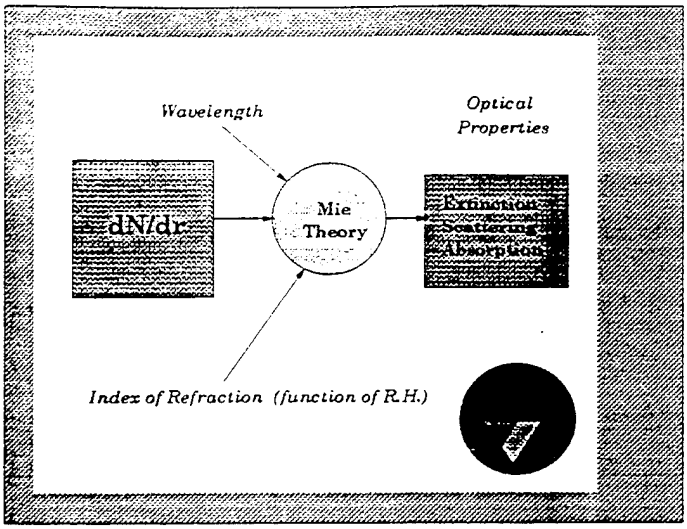
Pre Calculated Mie Integrals in NAM/NOVAM

$$\beta_{ext} = \frac{\pi}{1000} \int Q_{ext} \cdot \frac{dN}{dr} \cdot r^2 \cdot dr$$

$$\beta_{ext} = \frac{\pi}{1000} \int Q_{ext} \cdot \sum_{i=0}^{3} \frac{A_i}{f_i} \cdot \exp\left[-1 \cdot \left[\log_{10}\left(\frac{r}{C_i \cdot f_i}\right)\right]^2\right] \cdot r^2 \cdot dr$$

$$\beta_{ext} = \frac{\pi}{1000} \sum_{i=0}^{3} \frac{A_i}{f_i} \int Q_{ext} \cdot \exp\left[-1 \cdot \left[\log_{10}\left(\frac{r}{C_i f_i}\right)\right]^2\right] \cdot r^2 \cdot dr$$





Naval Oceanic Vertical Aerosol Model

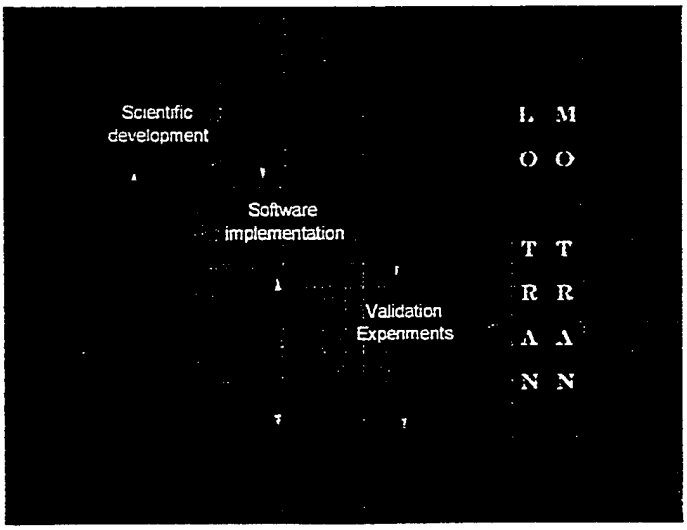
- Need for vertical structure of aerosol
- Use of NAM as a kernel
- Cooperation of NRL, NRaD, and NPS
- Identification of different marine structures
- Getting models which corresponded to these structures
- Software integration of the models

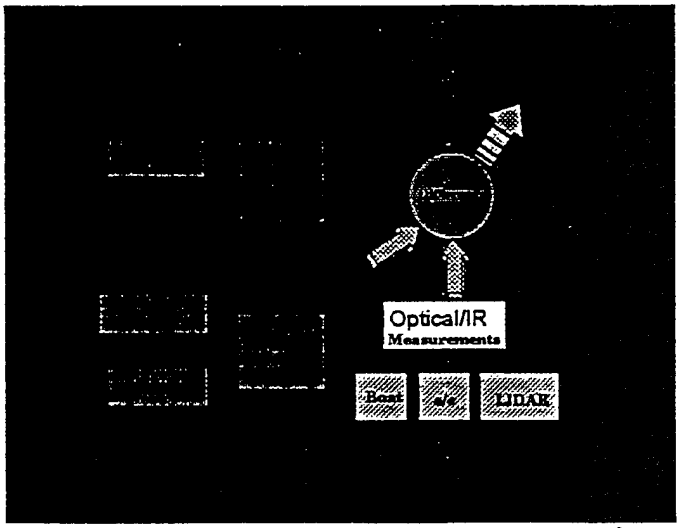
Assimulation of atmospheric structure into model

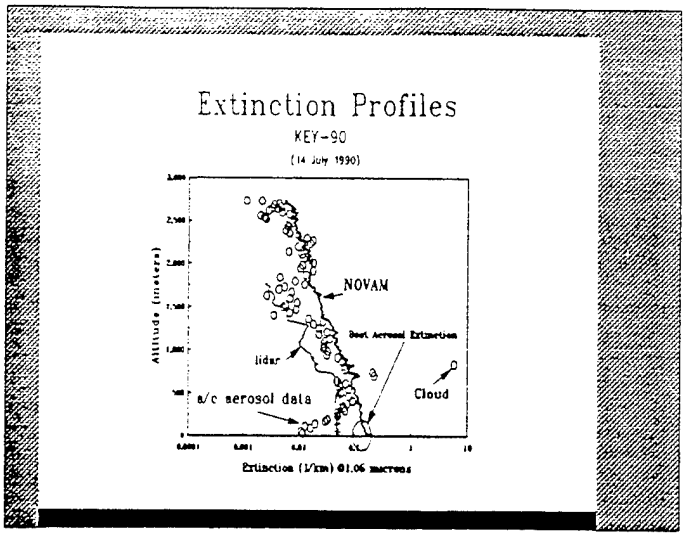
- manual approach
  - input hand calculated parameters from the profile into an input data file.
- semi automatic approach
  - stand alone aerosol program for the PC
  - computer makes a best guess, and then the user interactively adjusts curve to fit the data as needed
- fully automatic approach -- needed for a non interactive programs
  - a FORTRAN subroutine developed for LOWTRAN / MODTRAN

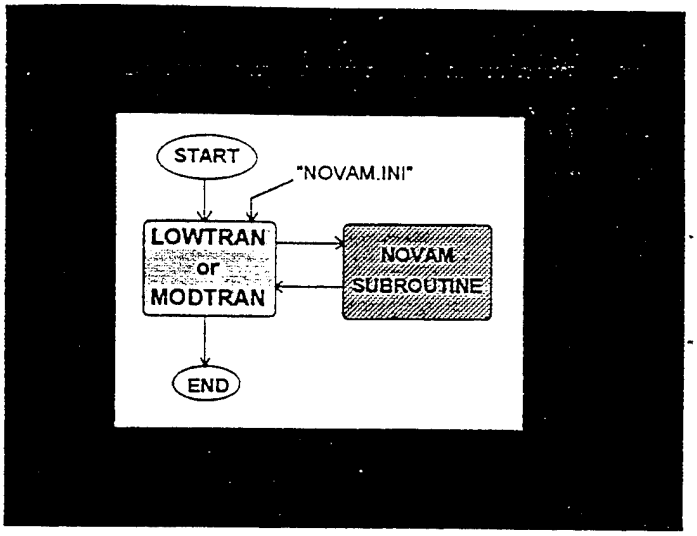
Testing of NOVAM in different geographical areas

- California coast - FIRE
- Tropical environment - KEY-90
- East coast - piggy back on IRAMMP 91 and IRAMMP 92 experiments
- Oregon coast - with NRL Blimp
- North Sea - MAPTIP experiment.









Limitations of NAM and NOVAM

- The near sea surface case
- Problems with the characterization of the continental air mass parameter based on simple measurements.
  - probablaly should be used in more open ocean regions
  - visibility used to characterize A1
  - other types of parameterization
  - development of the CAM

Areas of current thrusts

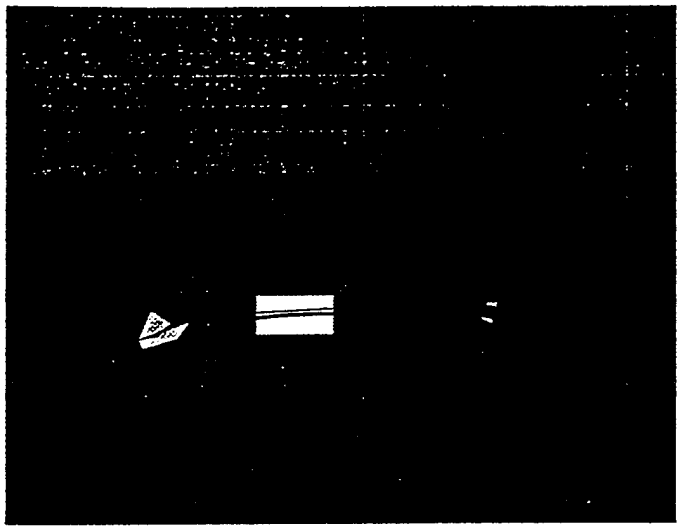
- NOVAM
- ANAM
- CAM

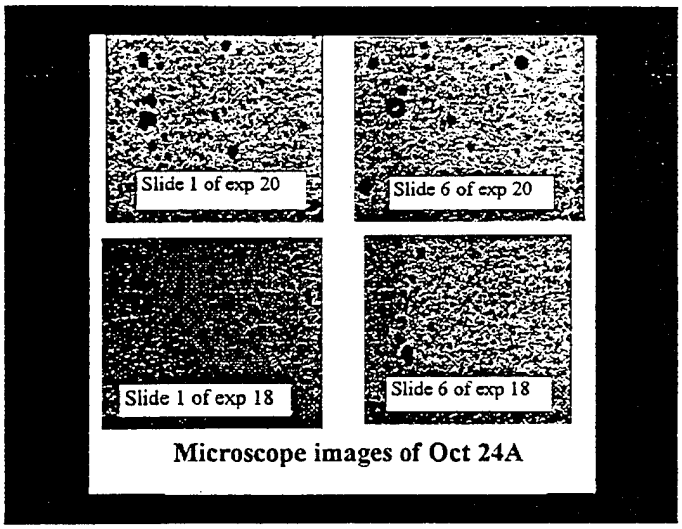
NOVAM

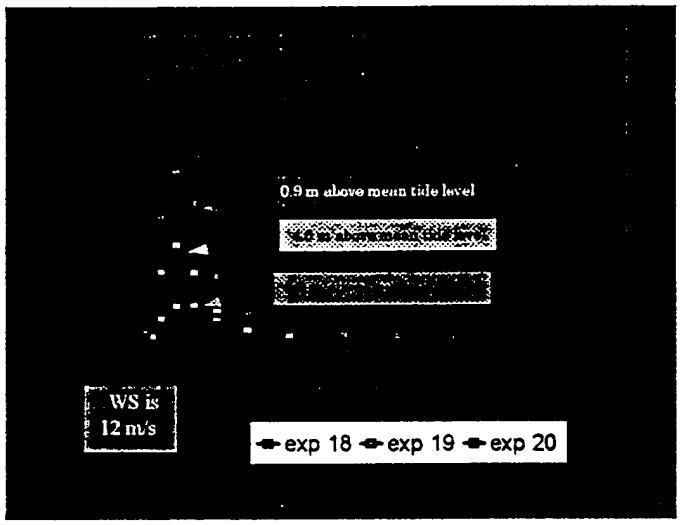
- default parameters (no meteorological profiles available)
  - Use of most likely profile for any MARSTEN square and at any month of the year

ANAM

- Interaction with 6.1 sea salt production mechanism people
- Issues in obtaining of data close to the sea surface
  - methods
    - Insitu techniques
    - Remote sensing techniques
    - transmissometer techniques
  - geometry of sea surface

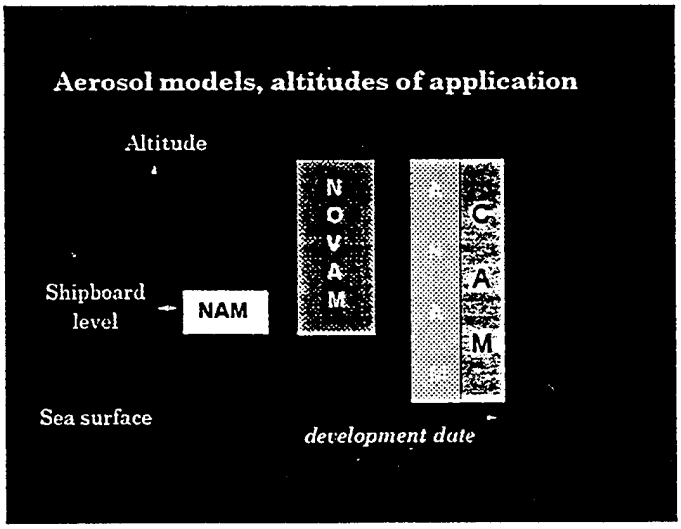






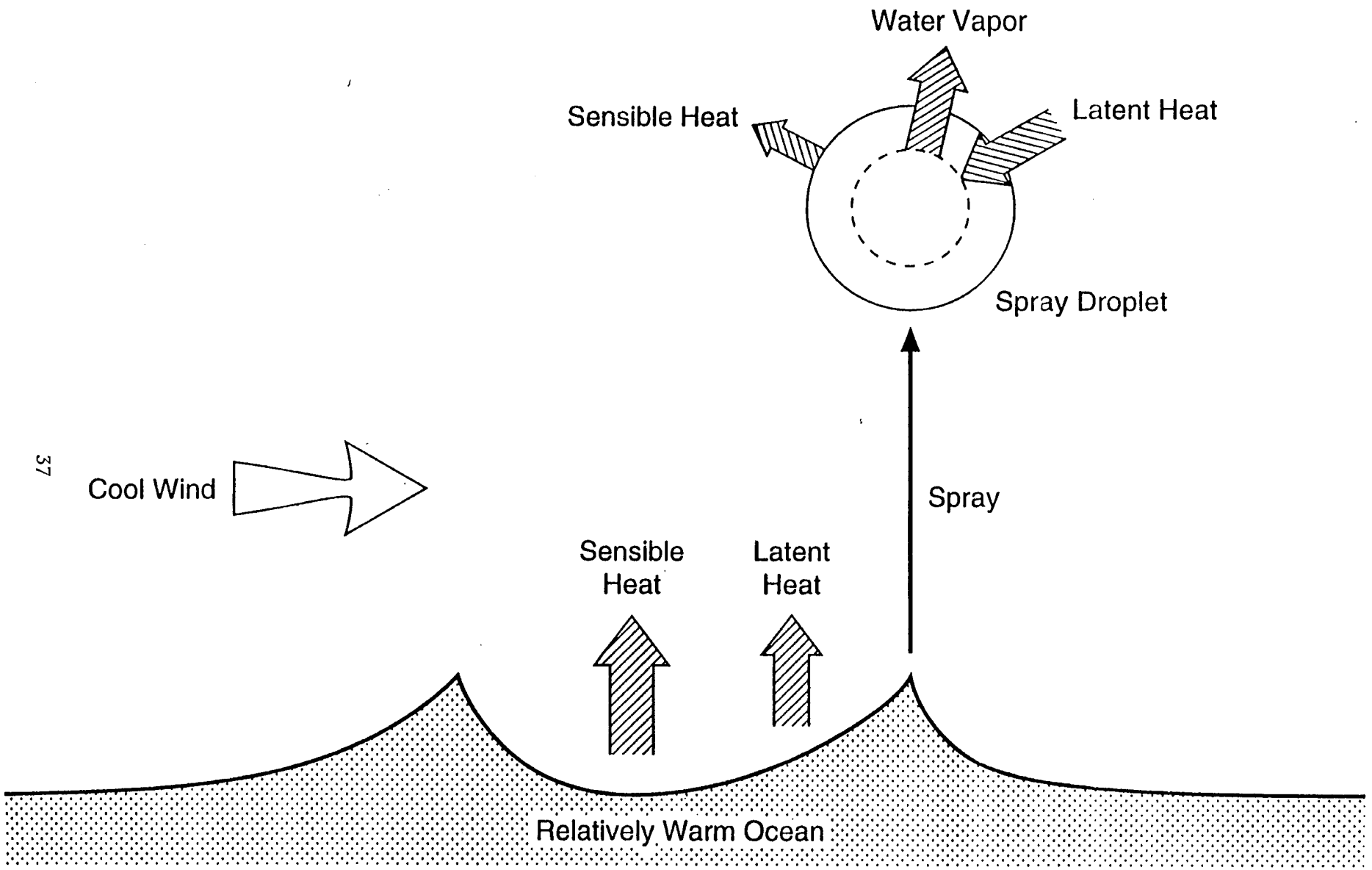
CAM

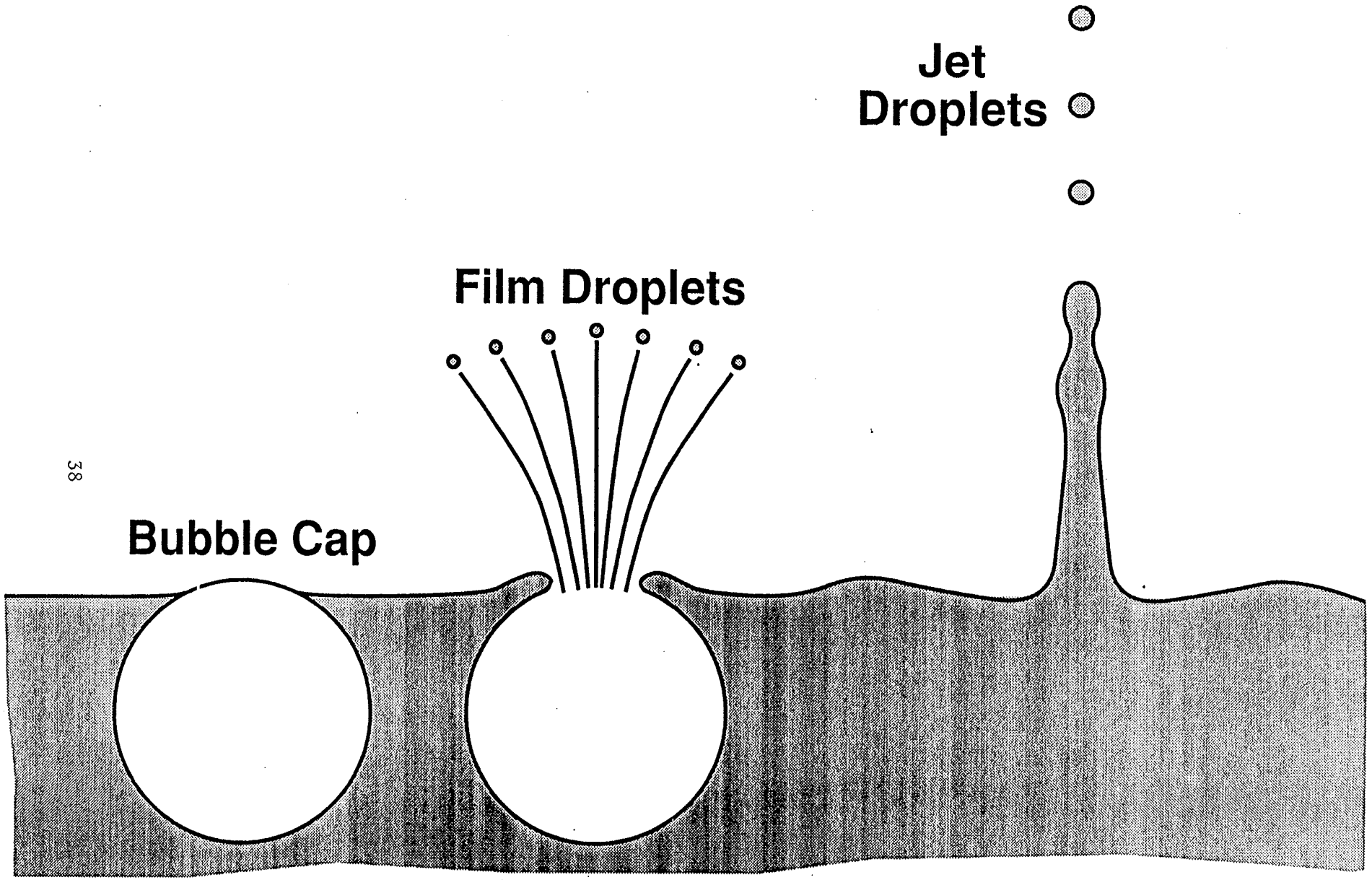
- Issues with the coastal aerosol modeling problem
  - Scope or scale of the model
  - characterization of anthropological aerosol sources
    - cities and industrial areas
    - battlefield effects - Army involvement
  - characterization of natural aerosol sources - sand & dust
  - what effect does surf line have on coastal aerosols
  - what is the effect of regional wind patterns on aerosol advection

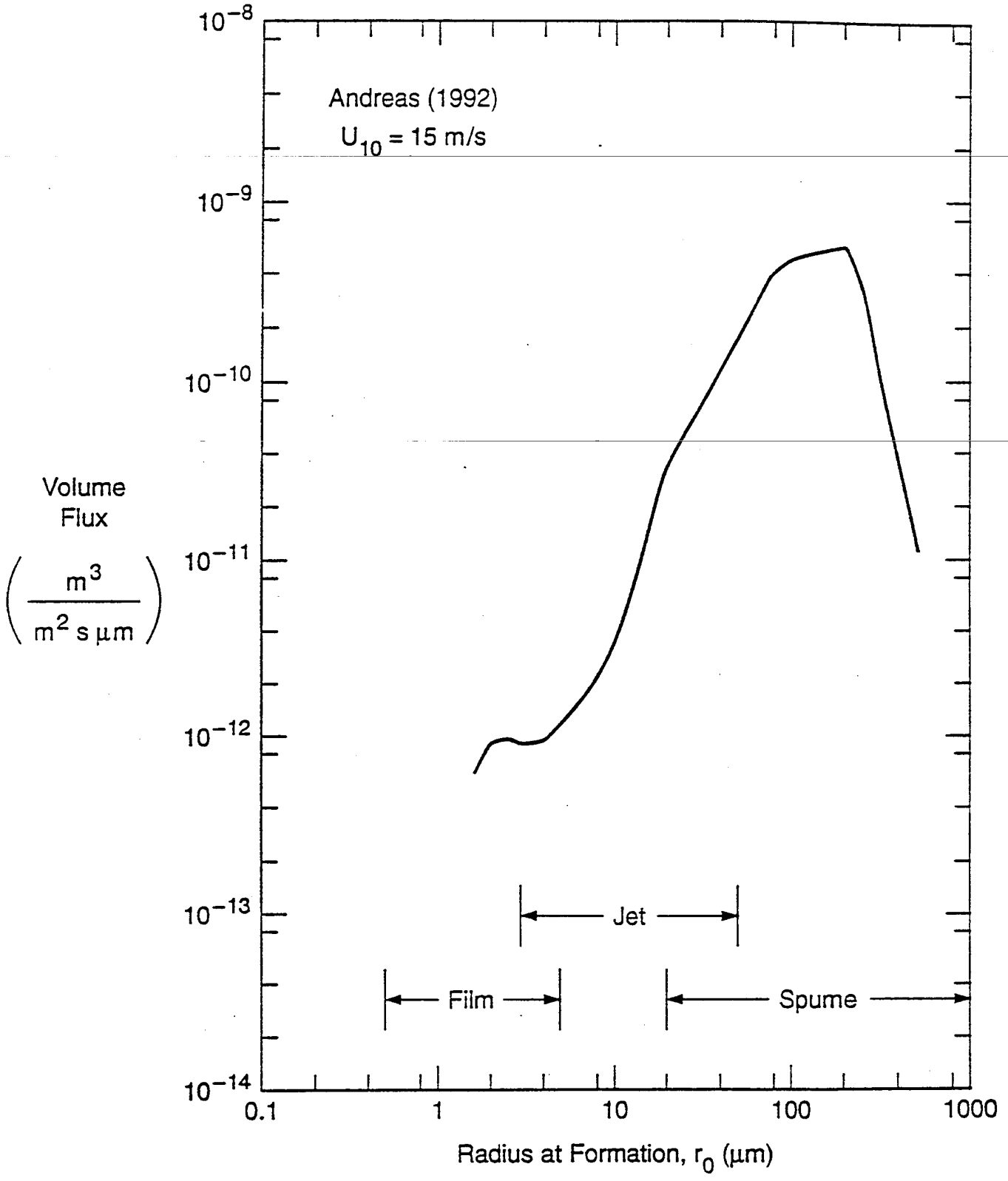


Surface Aerosol Sources

Dr. Edgar Andreas
US Army CRREL
Hannover New Hampshire


Water Vapor
Sensible Heat
Latent Heat
Spray Droplet
Cool Wind
Spray
Sensible Heat
Latent Heat
Relatively Warm Ocean


Jet
Droplets
Film Droplets
Bubble Cap


Andreas (1992)
U_{10} = 15 m/s
Volume Flux
$\left(\frac{m^3}{m^2\ s\ \mu m}\right)$
10^{-8}
10^{-9}
10^{-10}
10^{-11}
10^{-12}
10^{-13}
10^{-14}
Jet
Film
Spume
0.1
1
10
100
1000
Radius at Formation, r_0 (μm)

TIME CONSTANTS

τ_T, Temperature equilibrium:

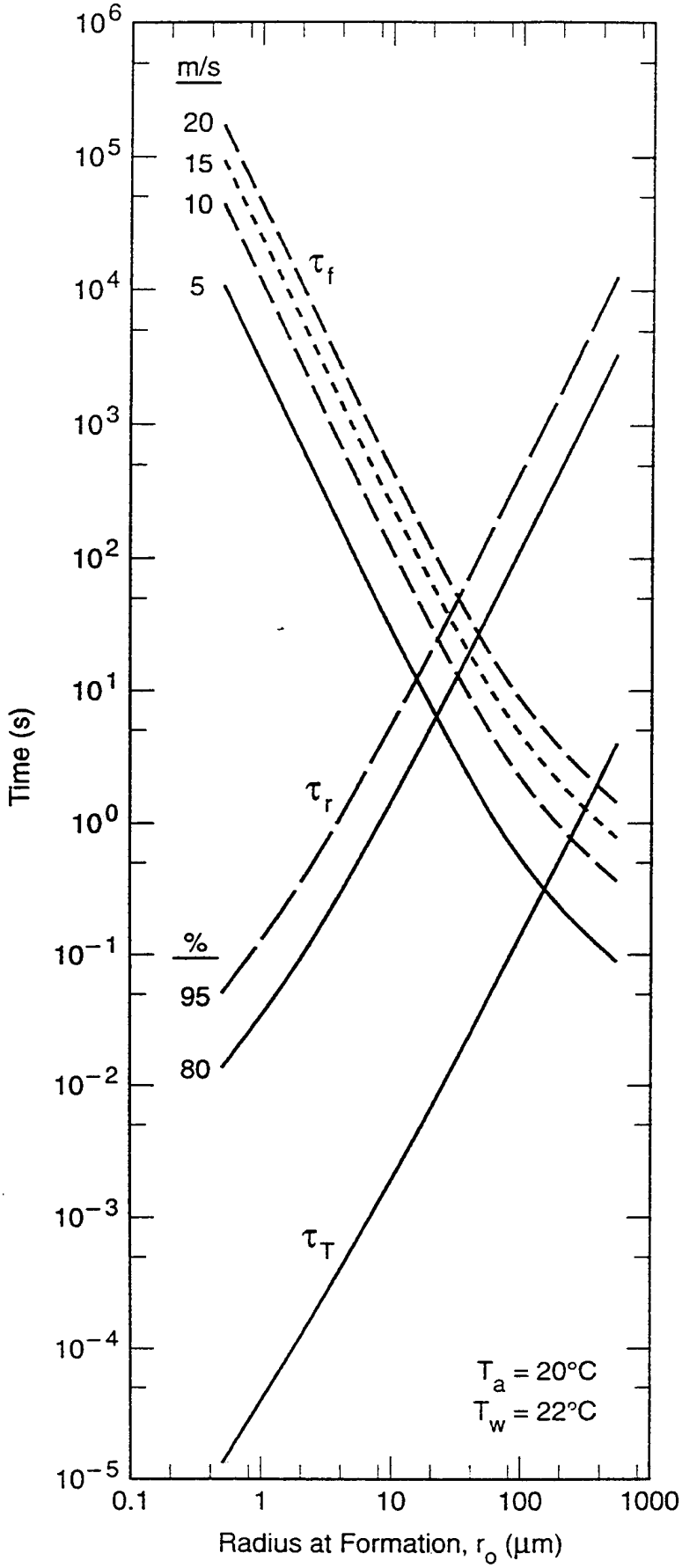
$$\frac{T(\tau_T) - T_{eq}}{T_w - T_{eq}} = e^{-1}$$

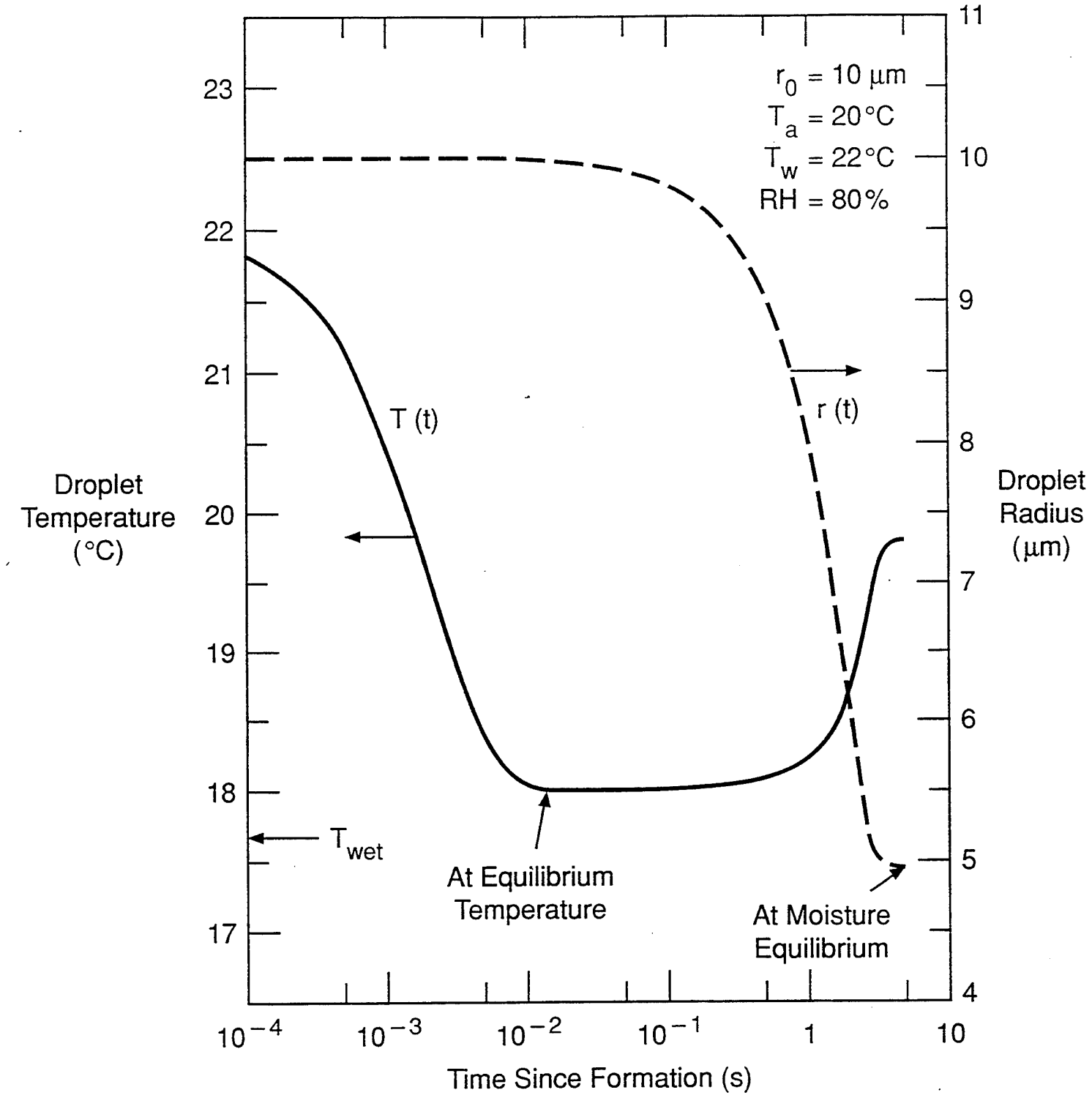
τ_r, Moisture (size) equilibrium:

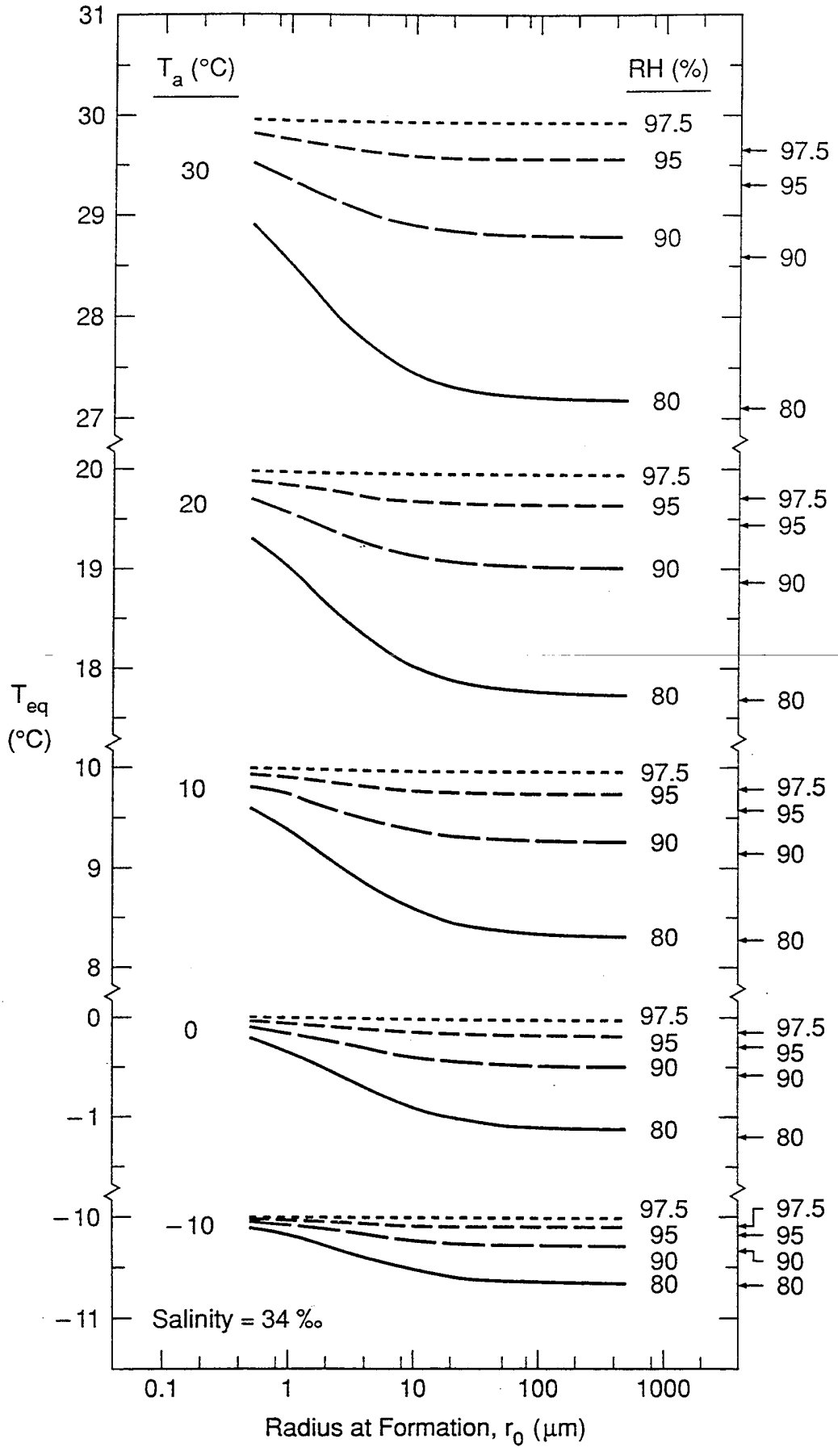
$$\frac{r(\tau_r) - r_{eq}}{r_o - r_{eq}} = e^{-1}$$

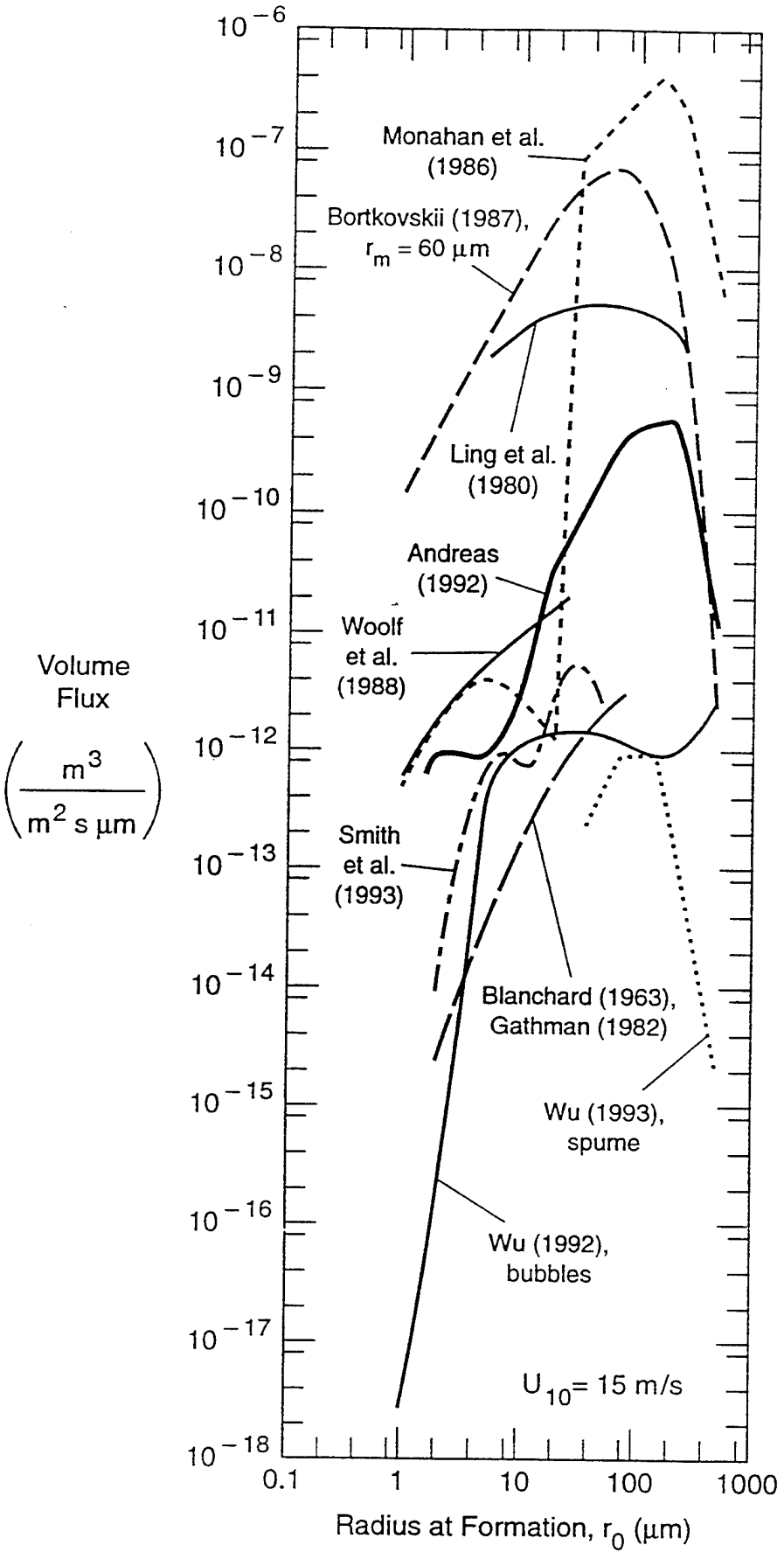
T_f, To fall from a height of $A_{1/3}$:

$$\tau_f = \frac{A_{1/3}}{u_f}$$


m/s
20
15
10
5
τ_f
τ_r
τ_T
%
95
80
T_a = 20°C
T_w = 22°C
Time (s)
Radius at Formation, r_o (μm)
10^6
10^5
10^4
10^3
10^2
10^1
10^0
10^{-1}
10^{-2}
10^{-3}
10^{-4}
10^{-5}
0.1
1
10
100
1000


r_0 = 10 μm
T_a = 20°C
T_w = 22°C
RH = 80%
T (t)
r (t)
T_{wet}
At Equilibrium Temperature
At Moisture Equilibrium
Droplet Temperature (°C)
Droplet Radius (μm)
Time Since Formation (s)
23
22
21
20
19
18
17
11
10
9
8
7
6
5
4
10^{-4}
10^{-3}
10^{-2}
10^{-1}
1
10


T_a (°C)
RH (%)
30
20
10
0
−10
97.5
95
90
80
T_{eq} (°C)
31
30
29
28
27
20
19
18
10
9
8
0
−1
−10
−11
Salinity = 34 ‰
0.1
1
10
100
1000
Radius at Formation, r_0 (μm)


Volume Flux
$\left(\frac{m^3}{m^2\ s\ \mu m}\right)$
10^{-6}
10^{-7}
10^{-8}
10^{-9}
10^{-10}
10^{-11}
10^{-12}
10^{-13}
10^{-14}
10^{-15}
10^{-16}
10^{-17}
10^{-18}
Monahan et al. (1986)
Bortkovskii (1987), r_m = 60 μm
Ling et al. (1980)
Andreas (1992)
Woolf et al. (1988)
Smith et al. (1993)
Blanchard (1963), Gathman (1982)
Wu (1993), spume
Wu (1992), bubbles
U_{10}= 15 m/s
0.1
1
10
100
1000
Radius at Formation, r_0 (μm)

SPRAY GENERATION FUNCTION/WHITECAPS/WIND

$$\frac{dF}{dr} \sim W \sim \dot{E} \sim \tau u_* = \rho u_*^3$$

where

$\frac{dF}{dr}$ = spray generation function (number m^{-2} s^{-1} μm^{-1})

W = fractional area of whitecap coverage

$\dot{E}$ = energy flux from the wind

τ = surface stress, ρu_*^2

u_* = friction velocity

TO FORM A SPUME DROPLET

Must change the free energy (ΔF),

$$\Delta F = -V(p_{in} - p_{out}) + \sigma \Omega$$

where

V = volume of droplet, $\frac{4}{3}\pi r^3$

p_{in} = pressure inside the droplet

p_{out} = pressure outside the droplet

σ = surface tension

Ω = surface area of droplet, $4\pi r^2$

r = droplet radius

From the Laplace equation,

$$p_{in} - p_{out} = \frac{2\sigma}{r}$$

Therefore,

$$\Delta F = \frac{1}{3}\sigma \Omega$$

SPRAY GENERATION FUNCTION/SPUME

Total rate of free energy change, all droplets,

$$\Delta\dot{F} = \frac{1}{3}\,\sigma\,\dot{\Omega}_t$$

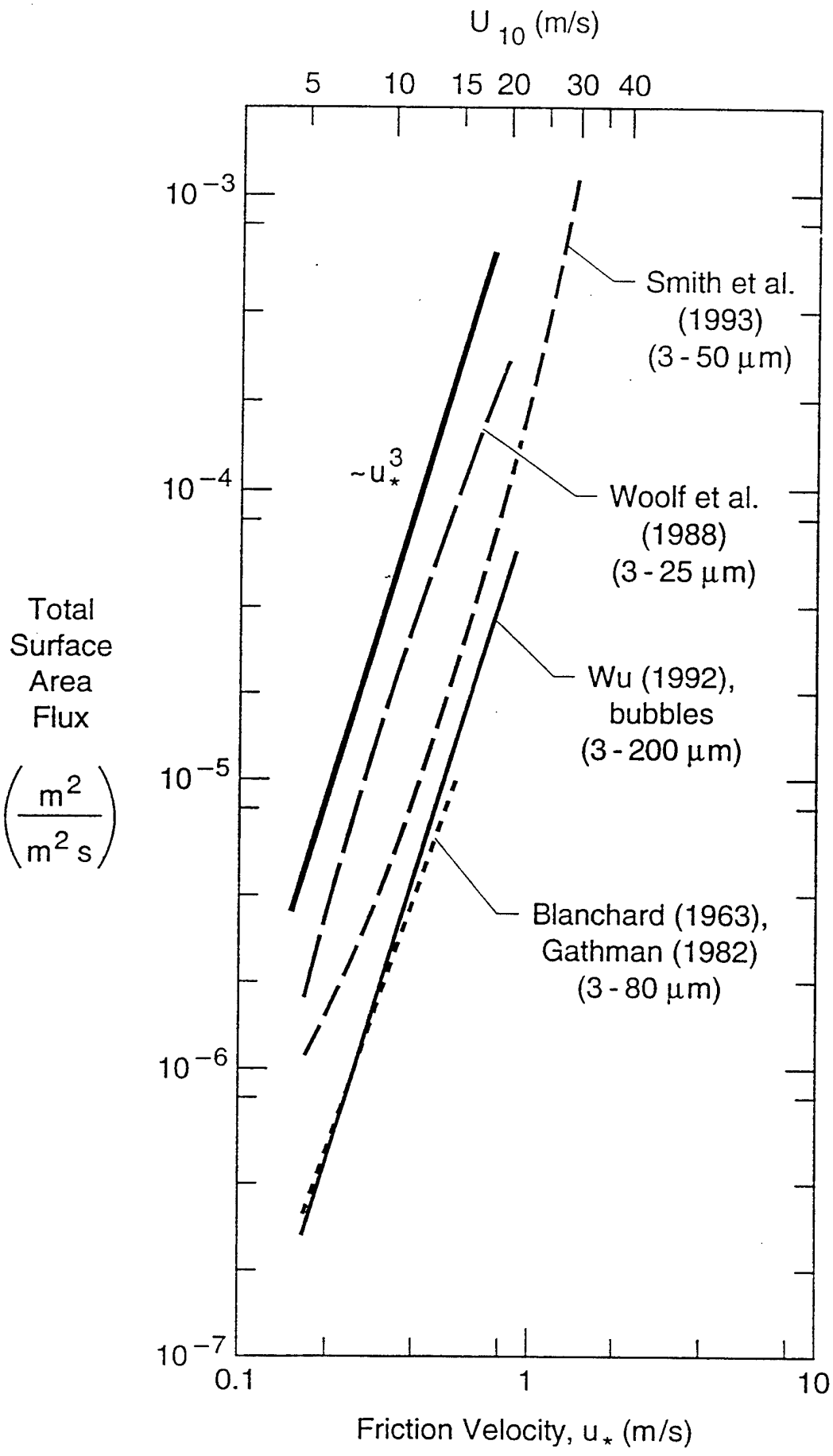
where the total rate of surface area formation is

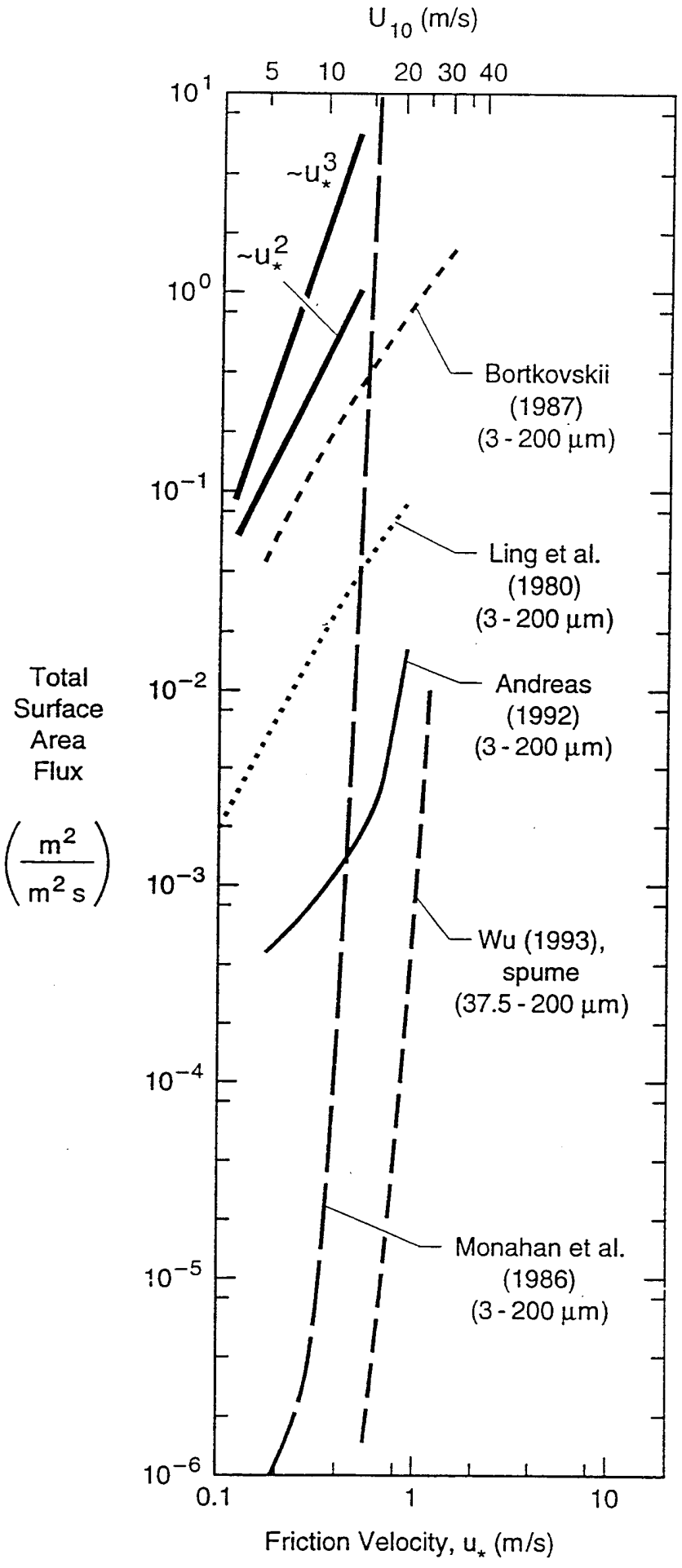
$$\dot{\Omega}_t = 4\pi\int_0^\infty r^2\left(\frac{dF}{dr}\right)dr$$

Units of $\Delta\dot{F}$ are W/m^2, the same as $\rho\,u_*^3$.

Therefore, hypothesize

$$\dot{\Omega}_t \sim \rho\,u_*^3$$


U 10 (m/s)
5
10
15
20
30
40
10 −3
10 −4
10 −5
10 −6
10 −7
~u*3
Smith et al.
(1993)
(3 - 50 μm)
Woolf et al.
(1988)
(3 - 25 μm)
Wu (1992),
bubbles
(3 - 200 μm)
Blanchard (1963),
Gathman (1982)
(3 - 80 μm)
Total
Surface
Area
Flux
(m2 / m2 s)
0.1
1
10
Friction Velocity, u* (m/s)


U_{10} (m/s)
5
10
20
30
40
10^1
10^0
10^{-1}
10^{-2}
10^{-3}
10^{-4}
10^{-5}
10^{-6}
$\sim u_*^3$
$\sim u_*^2$
Bortkovskii
(1987)
(3 - 200 μm)
Ling et al.
(1980)
(3 - 200 μm)
Andreas
(1992)
(3 - 200 μm)
Wu (1993),
spume
(37.5 - 200 μm)
Monahan et al.
(1986)
(3 - 200 μm)
Total
Surface
Area
Flux
$\left(\frac{m^2}{m^2\,s}\right)$
0.1
1
10
Friction Velocity, u_* (m/s)

ATMOSPHERIC AEROSOLS
PHYSICS / CHEMISTRY / TRANSFORMATIONS

William A. Hoppel, Code 7228
Aerosol and Cloud Physics Section
Remote Sensing Division
Naval Research Laboratory

OVERVIEW

1. IMPORTANCE / CONNECTION OF AEROSOLS TO NAVY AND OTHER DISCIPLINES

2. LIFE-CYCLE PROCESSES:

   Sources, Sinks, Transformations, and Transport

3. AEROSOL LIFETIMES

23 May 1994

IMPORTANCE / CONNECTION OF AEROSOLS TO OTHER DISCIPLINES

* **E-M EXTINCTION / E-O SYSTEMS**

* **REMOTE SENSING**

  →Extinction (Detrimental effect)
  →Lidar (Aerosol provides backscatter signal)

* **CLOUD AND FOG MICROPHYSICS**

  →Aerosols provide nucleation sites (CCN)

* **CLIMATE**

  →Direct effect on radiation transfer
  →Indirect effect - cloud albedo

* **ATMOSPHERIC CHEMISTRY**

  →Homogeneous - Gas-phase reaction products removed by aerosols
  →Heterogeneous reactions on aerosols
  →Liquid-phase oxidation in cloud droplets

* **ATMOSPHERIC ELECTRICITY**

  →Aerosols are recombination centers for ions

$$\frac{S(\lambda,x)}{S_o(\lambda)} = \text{Exp}\left[-\int \beta_{ex}(\lambda,\bar{x})d\bar{x}\right]$$

WHERE

$$\beta_{ex}(\lambda,\bar{x}) = \int \pi r^2 Q_{ex}(r,\lambda) n(r,\bar{x}) dr$$

$Q_{ex} \equiv$ MIE EXTINCTION EFFICIENCY

$n(r) \equiv$ AEROSOL SIZE DISTRIBUTION

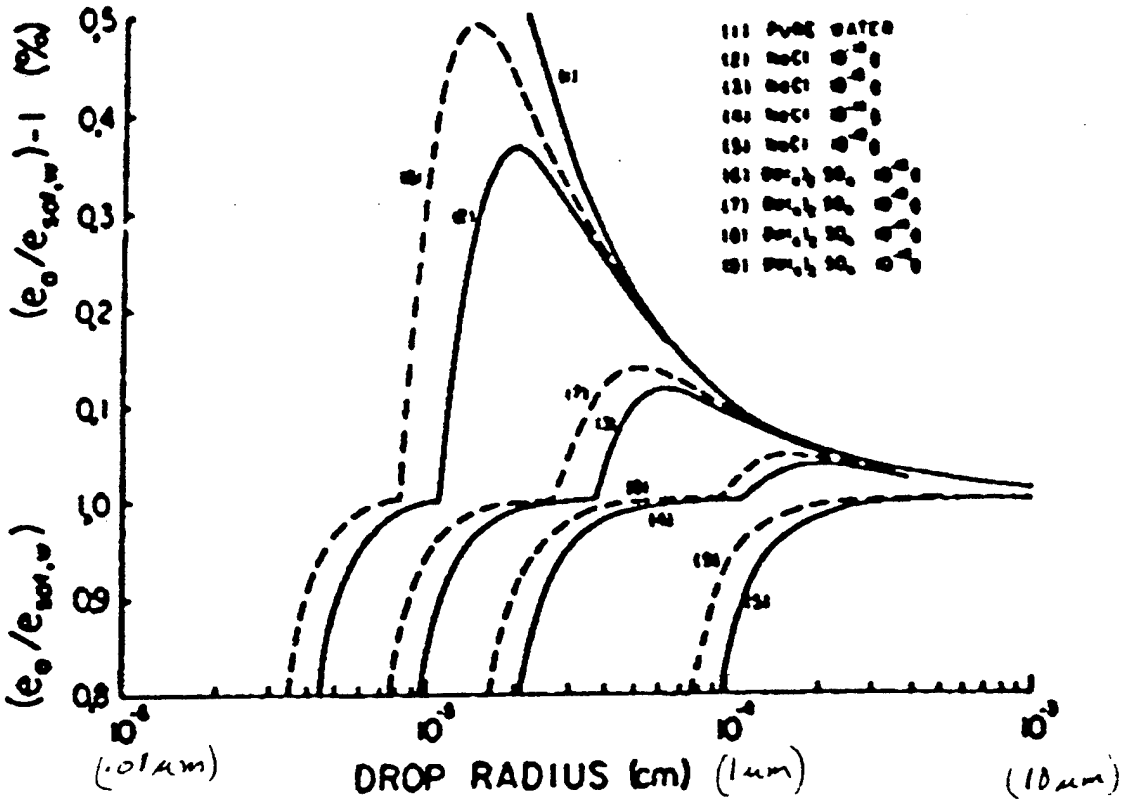
LIDAR EQUATION

$$P(R) \propto \frac{\beta_\pi(\lambda,R) \bullet EXP[-2 \int \beta_{ex}(\lambda,R)dx]}{R^2}$$

$$\beta_\pi(\lambda, R) = \int \pi r^2 Q_\pi(r, \lambda, m)\, n(r, R) dr$$

$$\beta_{ex}(\lambda, R) = \int \pi r^2 Q_{ex}(r, \lambda, m)\, n(r, R) dr$$

Critical Supersaturation Spectrum n(S) is Calculated from the Size Distribution n(r)



$$S = K r^{-3/2}$$

$$n(S) = -\frac{2}{3}\frac{r^{5/2}}{K} n(r).$$

CLIMATE CONNECTION

Warming resulting from greenhouse gases accumulated since 1850 is about +2.5 W/m^2

1. DIRECT EFFECT OF AEROSOLS (EST. -0.3 TO -1.5 W/m^2)

To calculate radiative transfer need:

$$\tau = \int \beta_{ex}(z)dz \quad \text{Optical depth}$$

$$\omega = \frac{\beta_{scat}}{\beta_{scat}+\beta_{abs}} \quad \text{Single scattering albedo}$$

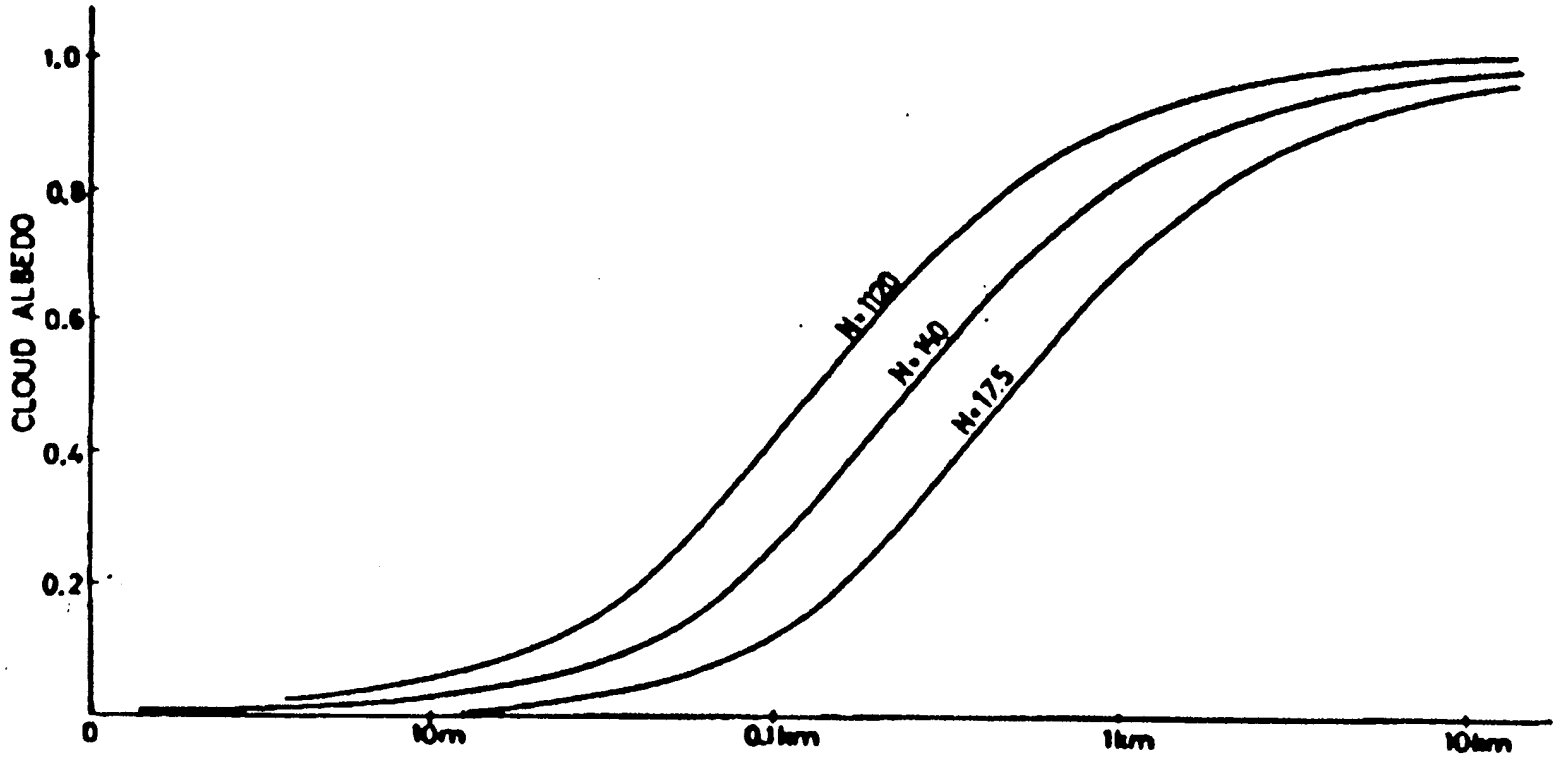
$$g = \frac{\pi \int r^2 Q_{scat}(r,\lambda,m)\, g(r,\lambda)\, n(r)\, dr}{\beta_{scat}} \quad \text{Bulk Asymmetry Factor}$$

2. INDIRECT EFFECT (Est. -0.5 to -2 W/m^2)
(TOA cloud short wave forcing 50 to 70 W/m^2; total cloud forcing 17 to 30 W/m^2)

* Change in cloud albedo due to increase in CCN concentration.

* Dramatic example of Navy Interest - Cloud tracks.

Effect of CCN Concentrations on Cloud Reflectivity



Cloud Thickness

EXAMPLES OF CONNECTION WITH CHEMISTRY

HOMOGENEOUS CHEMISTRY

If a trace gas is oxidized to a condensible species X at a rate Q then:

$$\frac{d[X]}{dt} = Q - 4\pi[X]\int r\,D'(r)[1 - \frac{[X_s(r)]}{[X]}]\,n(r)dr$$

Condensation causes particle growth

$$\frac{dr}{dt} = \frac{D'(r)\,m}{\rho_a r}\{1 - \frac{[X_s(r)]}{[X]}\}\,[X]$$

Particle growth causes change in the size distribution

$$\frac{\partial n(r)}{\partial t} = \frac{\partial}{\partial r}[n(r)\frac{dr}{dt}]$$

HETEROGENEOUS CHEMISTRY

Example:

$$CaCO_3 + H_2O + SO_2 + O_3 \longrightarrow CaSO_4 + H_2O + CO_2$$

Requires O_3 and RH above delequescent point

LIQUID PHASE CHEMISTRY IN CLOUD DROPLETS

CONNECTION TO ATMOSPHERIC ELECTRICITY

Aerosol particles act as centers of recombination for atmospheric ions - more important than ion-ion recombination.

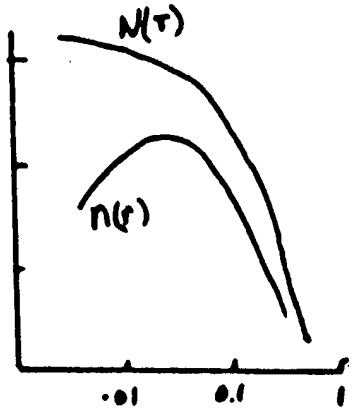
If N is the polar ion concentrations and q the ionization rate, then the ion balance is given by:

$$q = \alpha N^2 + \eta_{eff} Z N$$

where the effective ion aerosol attachment coefficient is

$$\eta_{eff} = \frac{\int \eta(r)\, n(r)\, dr}{Z} \quad \text{and} \quad Z = \int n(r)\, dr$$

N(r) = ***Cumulative Size Distribution***



$$n(r) \equiv \frac{dN(r)}{dr}$$ Differential Size Distribution

$$n'(r) \equiv \frac{dN(r)}{d[\log(r/r_0)]}$$ Logrithmic Size Distribution

$$\frac{dV(r)}{dr} \equiv \frac{4}{3}\pi r^3 \frac{dN}{dr}$$ Volumetric Size Distribution

LIFE-CYCLE PROCESSES

I. SOURCES

Primary Particles / Direct Injection

Secondary - *In situ* Formation from Reaction Products

II. SINKS

Surface Deposition

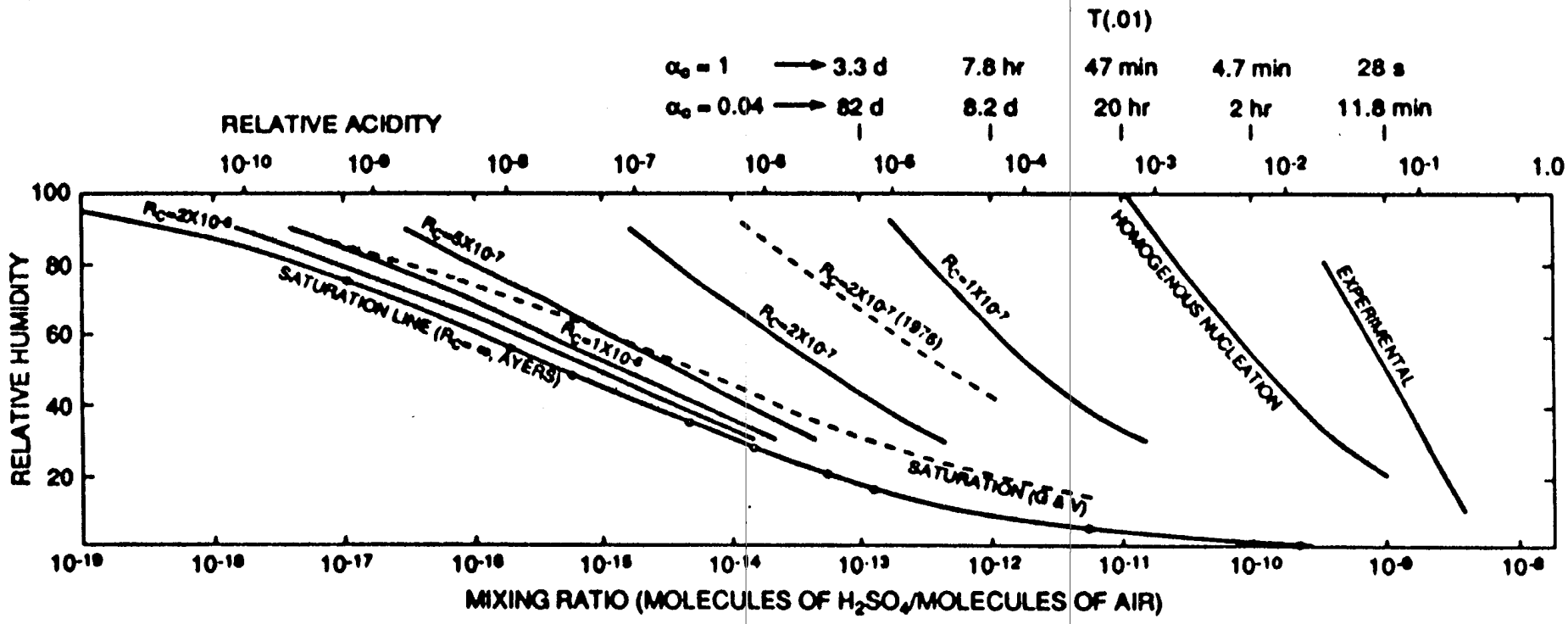
Precipitation Scavenging
- -Nucleation scavenging
- -Interstitial scavenging
- -Gravitational scavenging in and below cloud

III. TRANSFORMATIONS

Coagulation
Growth by Condensation of Reaction Products
Swelling by up-take of Water / changing RH
Cloud Processing
Heterogeneous Chemistry

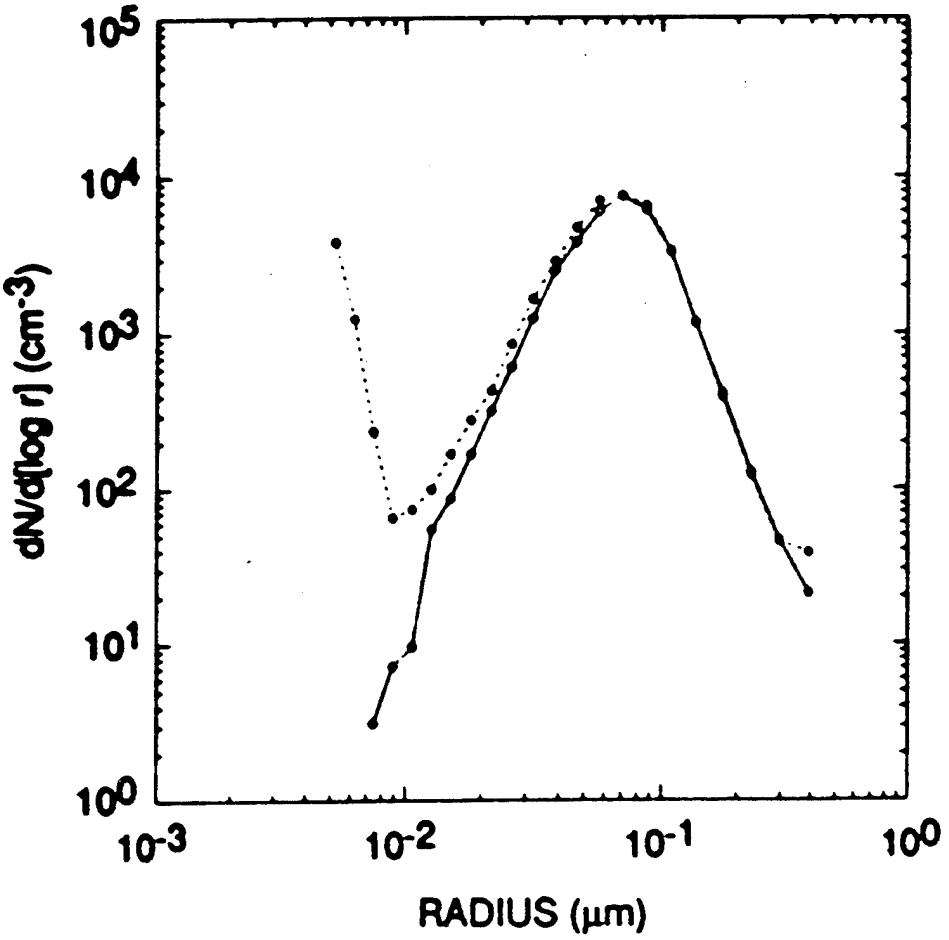
IV. TRANSPORT

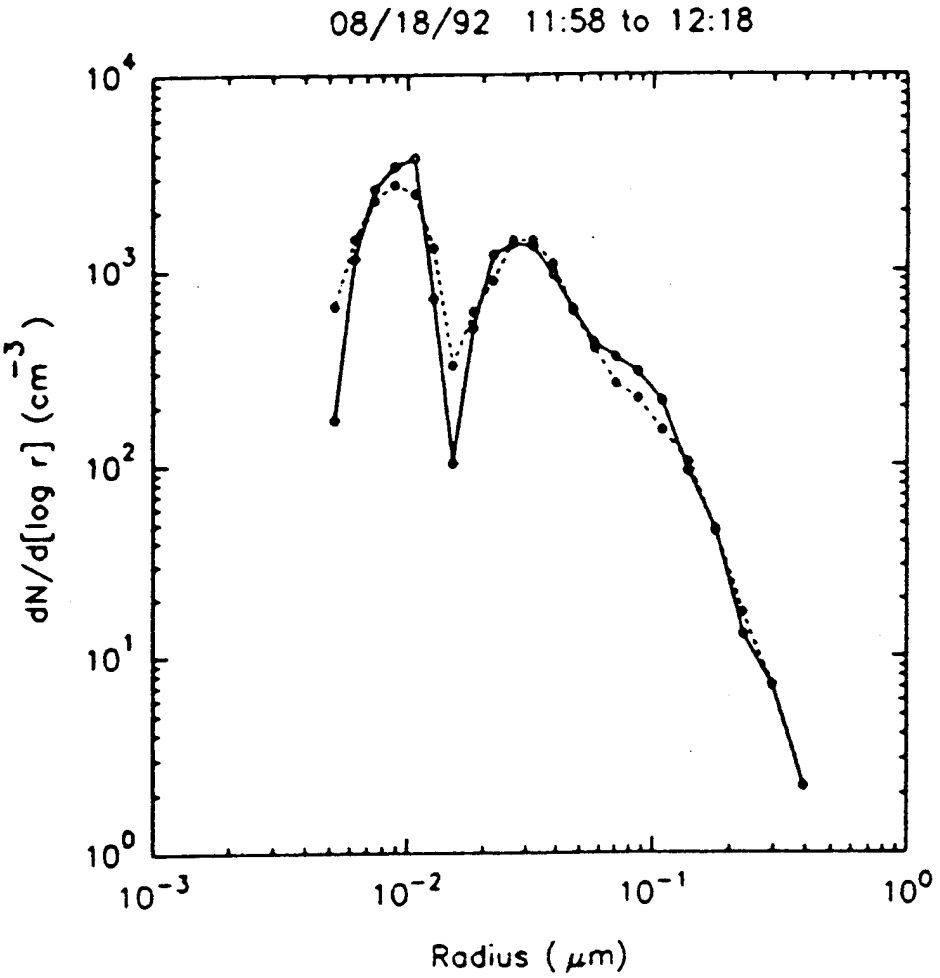
Advection
Vertical Mixing & Entrainment across Top of MBL

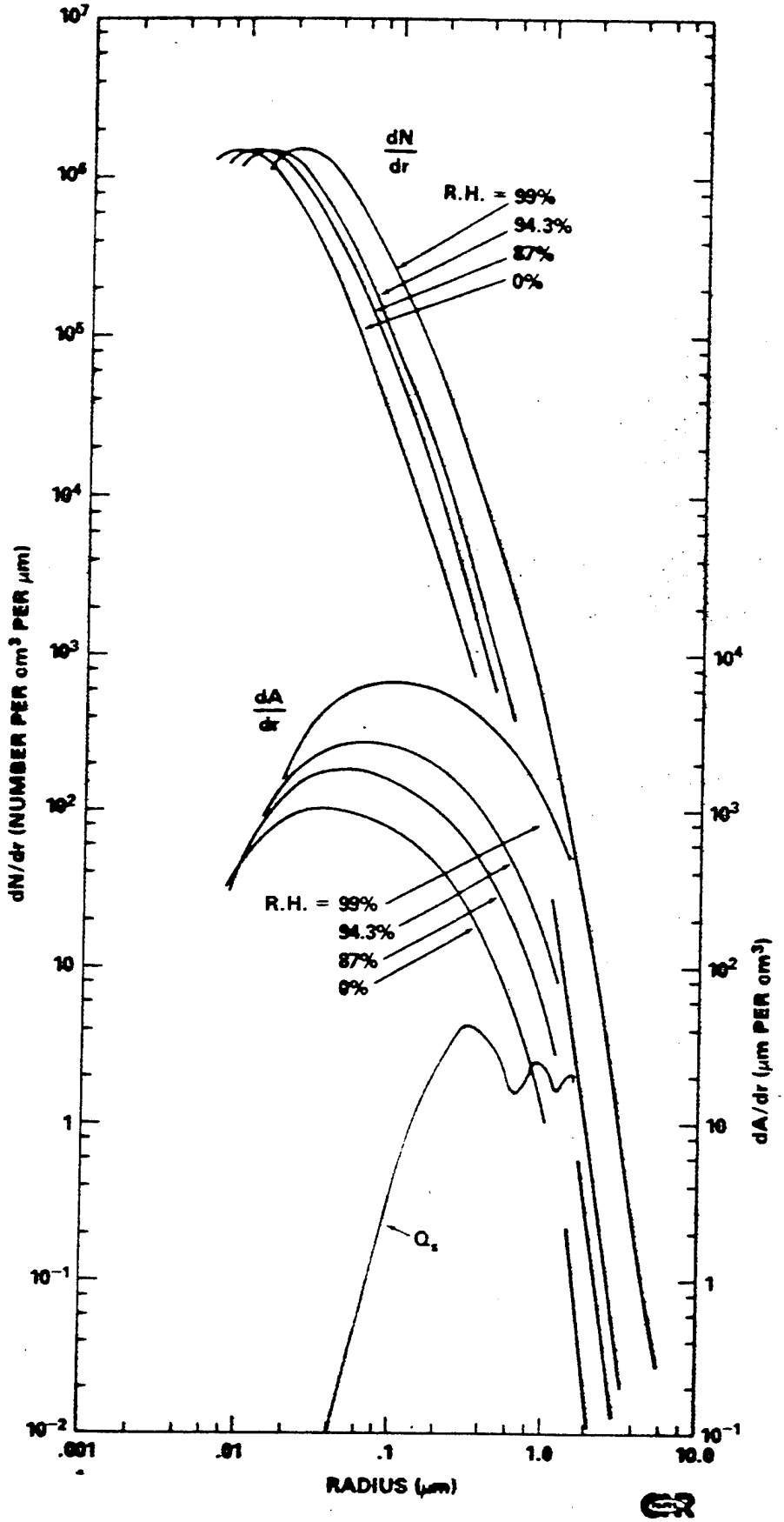

T(.01)
α_c = 1 → 3.3 d 7.8 hr 47 min 4.7 min 28 s
α_c = 0.04 → 82 d 8.2 d 20 hr 2 hr 11.8 min
RELATIVE ACIDITY
10-10 10-9 10-8 10-7 10-6 10-5 10-4 10-3 10-2 10-1 1.0
100
80
60
40
20
RELATIVE HUMIDITY
Rc=2X10-6
Rc=5X10-7
Rc=1X10-6
SATURATION LINE (Rc = ∞, AYERS)
Rc=2X10-7
Rc=2X10-7 (1976)
Rc=1X10-7
HOMOGENEOUS NUCLEATION
EXPERIMENTAL
SATURATION (δ = 0)
10-19 10-18 10-17 10-16 10-15 10-14 10-13 10-12 10-11 10-10 10-9 10-8
MIXING RATIO (MOLECULES OF H_2SO_4/MOLECULES OF AIR)

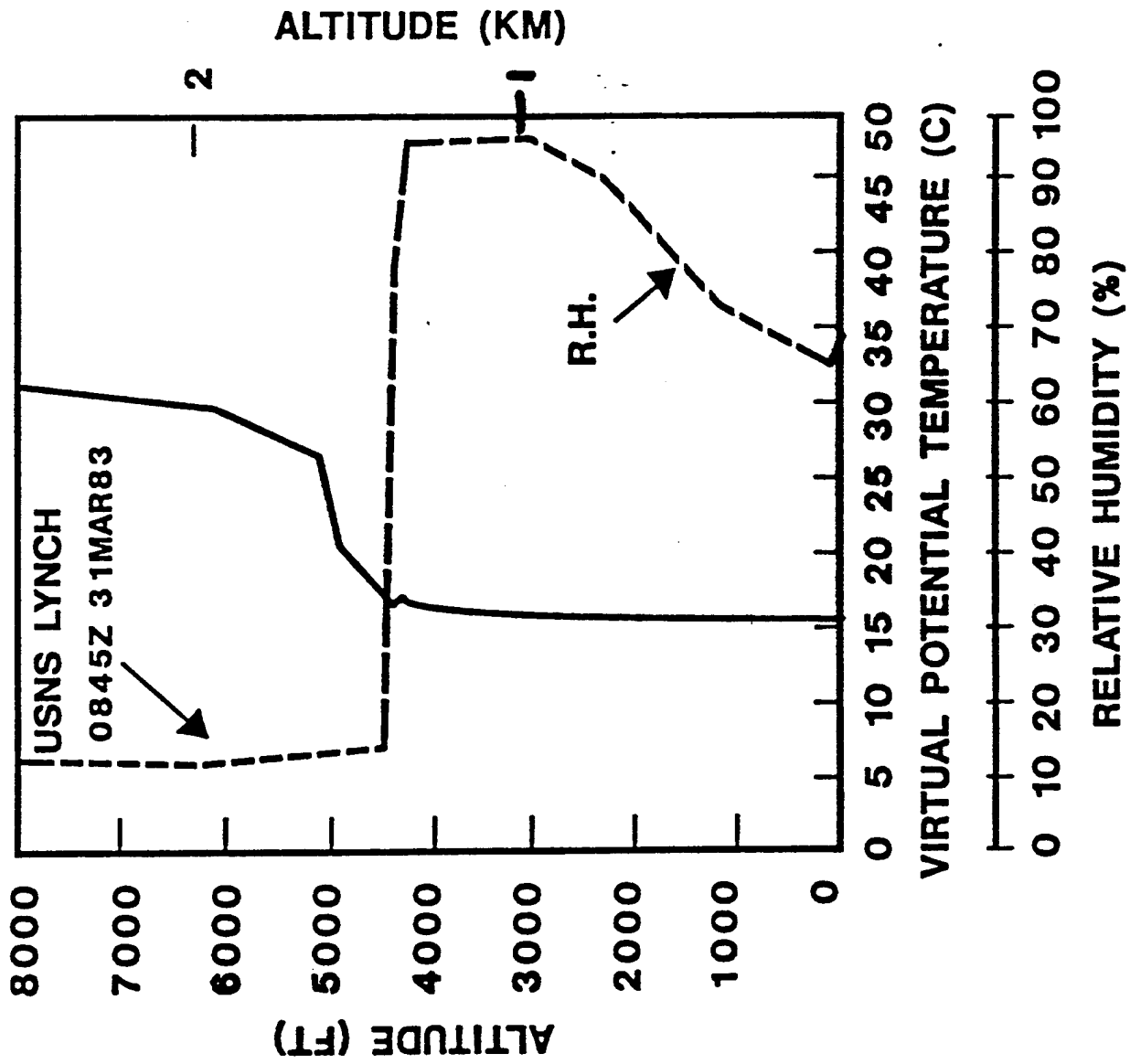
Two Size distribution taken in clear air about 1.5 hours apart both at an altitude of 600 ~~m~~ ft. The large numbers of small particles indicate a nucleation event is occurring.

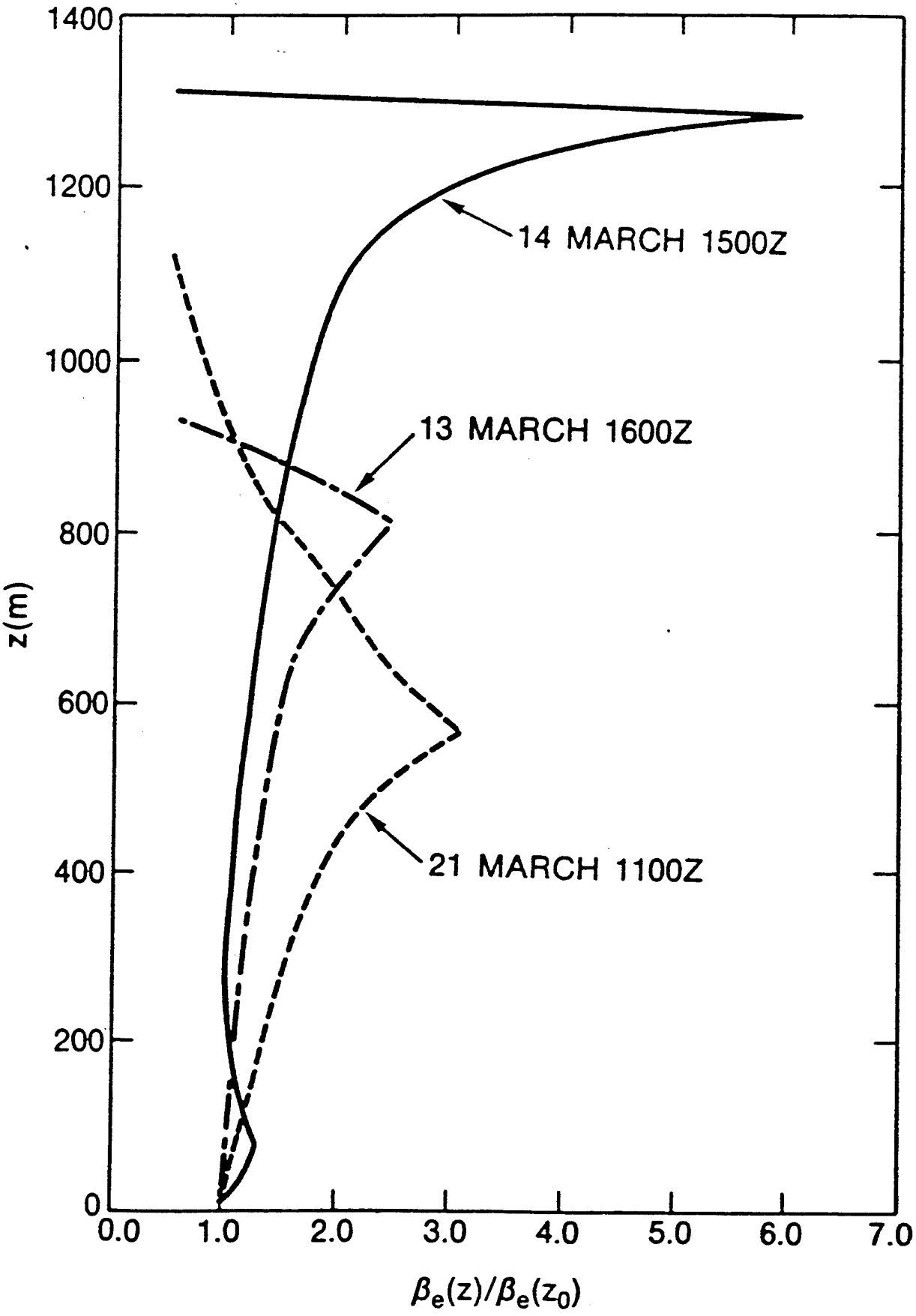
8/12/92

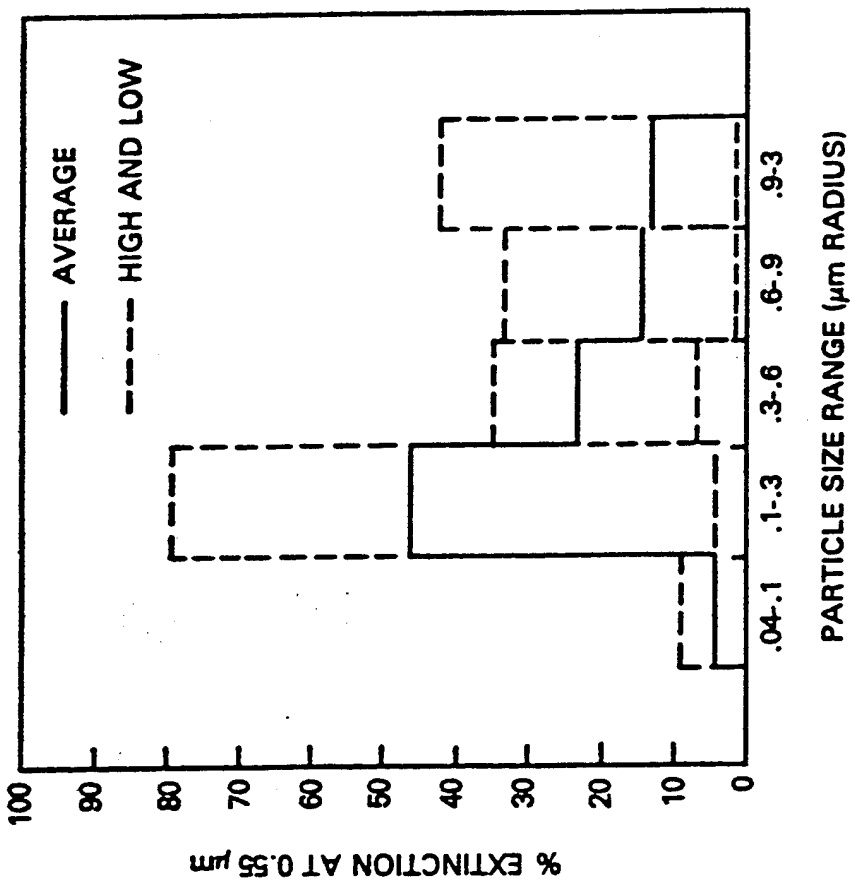


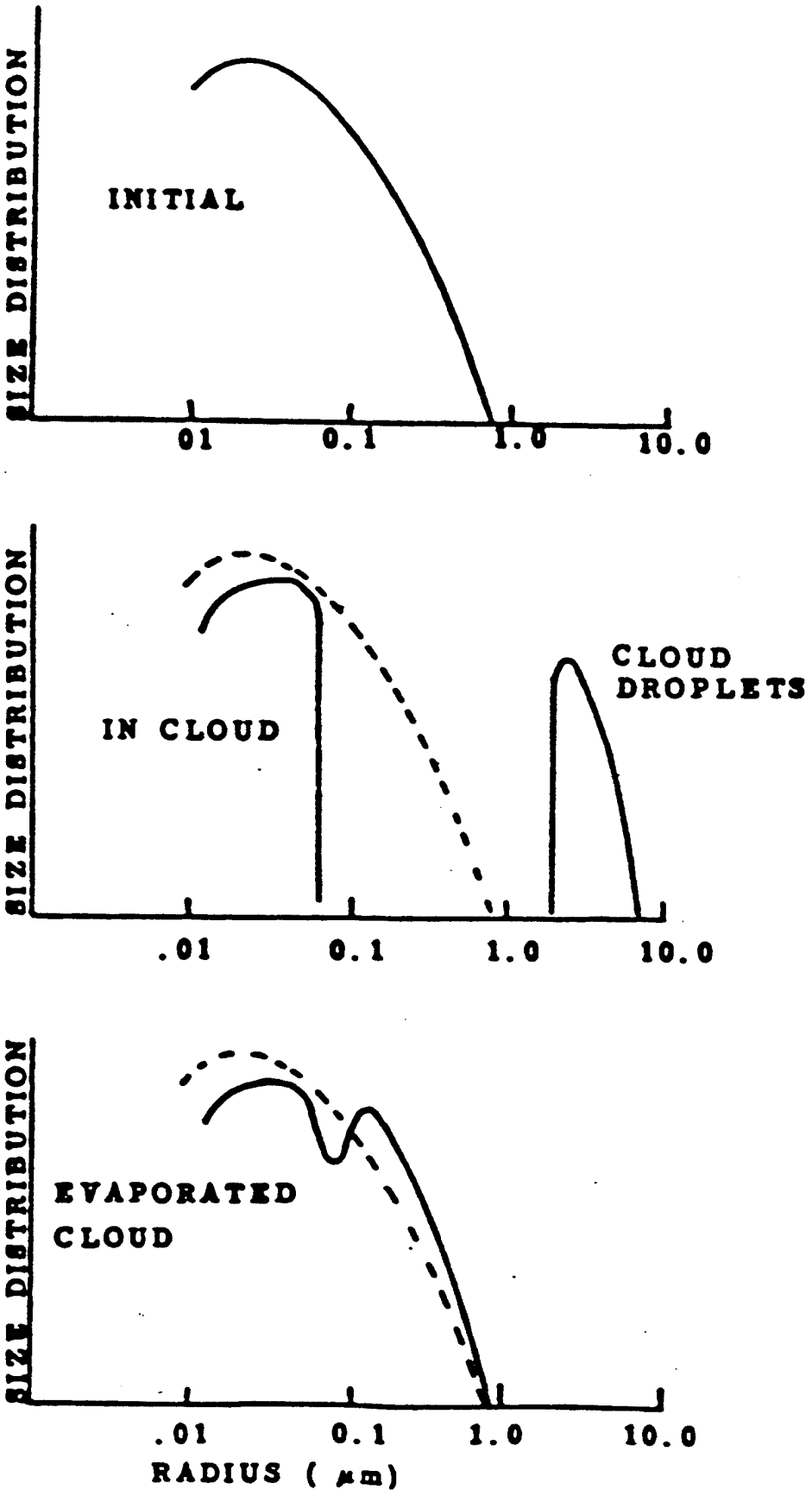

08/18/92 11:58 to 12:18
dN/d[log r] (cm^{-3})
10^4
10^3
10^2
10^1
10^0
10^{-3}
10^{-2}
10^{-1}
10^0
Radius (μm)

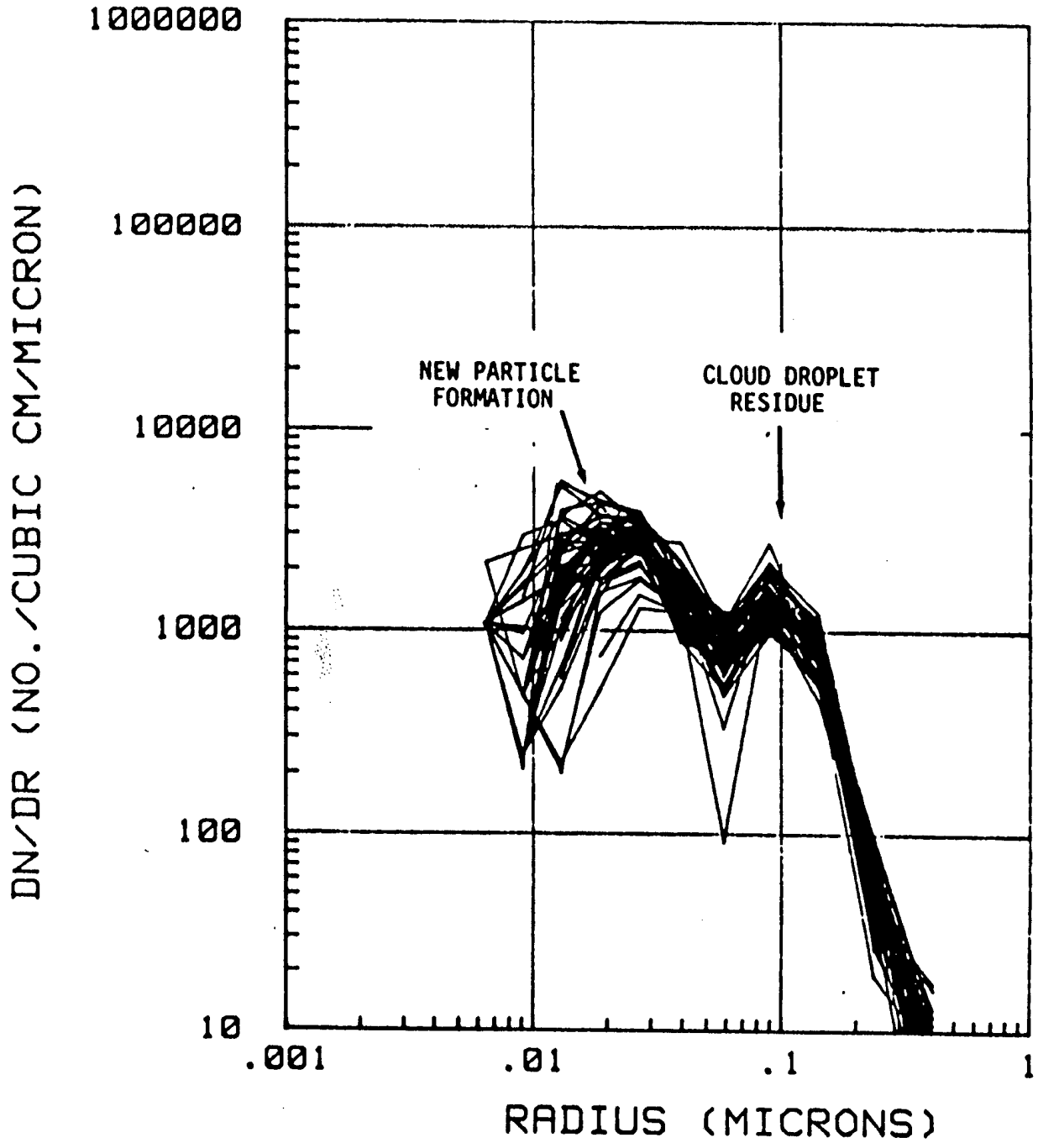

dN/dr (NUMBER PER cm³ PER μm)
dA/dr (μm PER cm²)
RADIUS (μm)
dN/dr
R.H. = 99%
94.3%
87%
0%
dA/dr
R.H. = 99%
94.3%
87%
0%
Qₑ
10⁷
10⁶
10⁵
10⁴
10³
10²
10
1
10⁻¹
10⁻²
10⁴
10³
10²
10
1
10⁻¹
.001
.01
.1
1.0
10.0

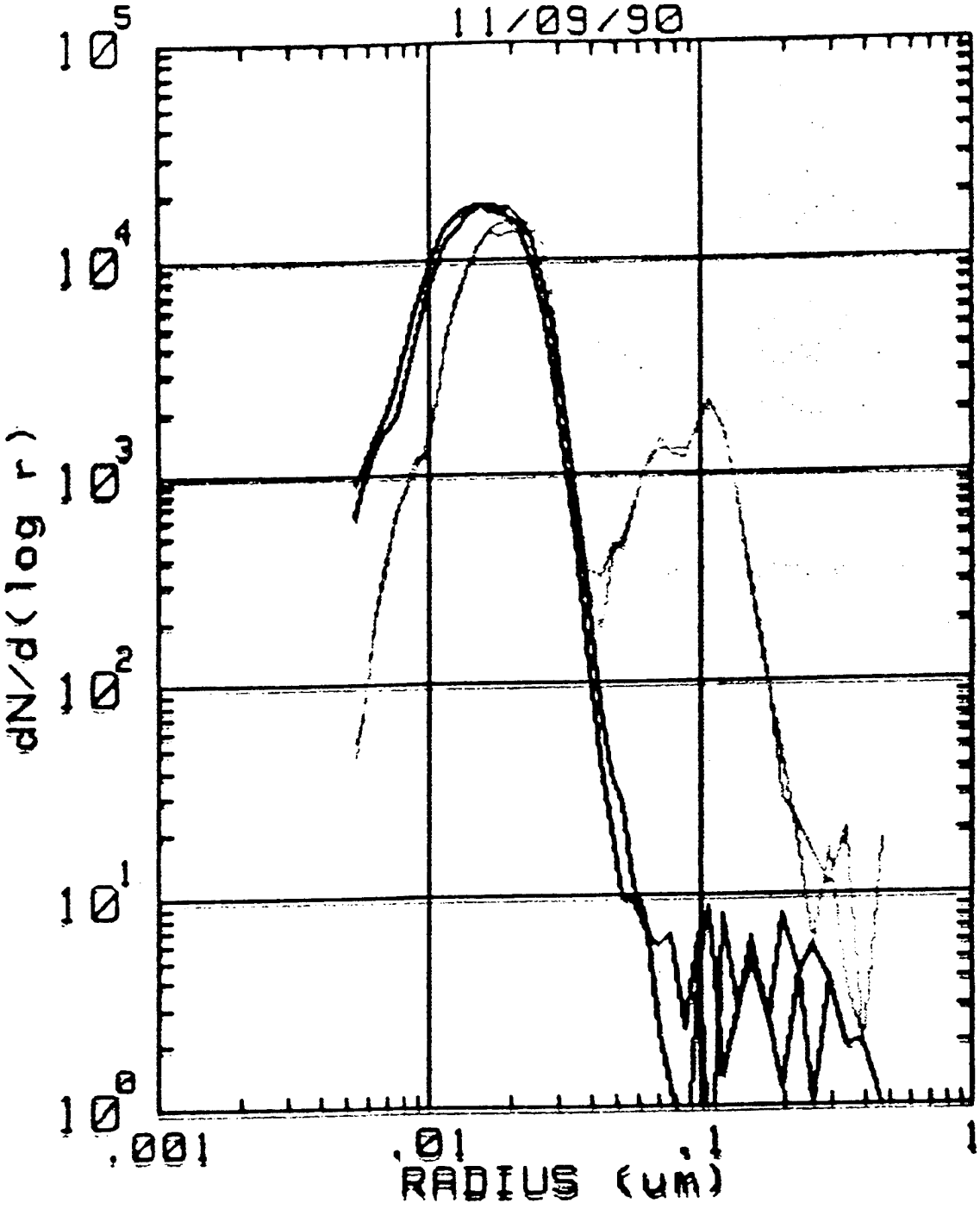

ALTITUDE (KM)
2
1
USNS LYNCH
0845Z 31MAR83
R.H.
8000
7000
6000
5000
4000
3000
2000
1000
0
ALTITUDE (FT)
0 5 10 15 20 25 30 35 40 45 50
VIRTUAL POTENTIAL TEMPERATURE (C)
0 10 20 30 40 50 60 70 80 90 100
RELATIVE HUMIDITY (%)

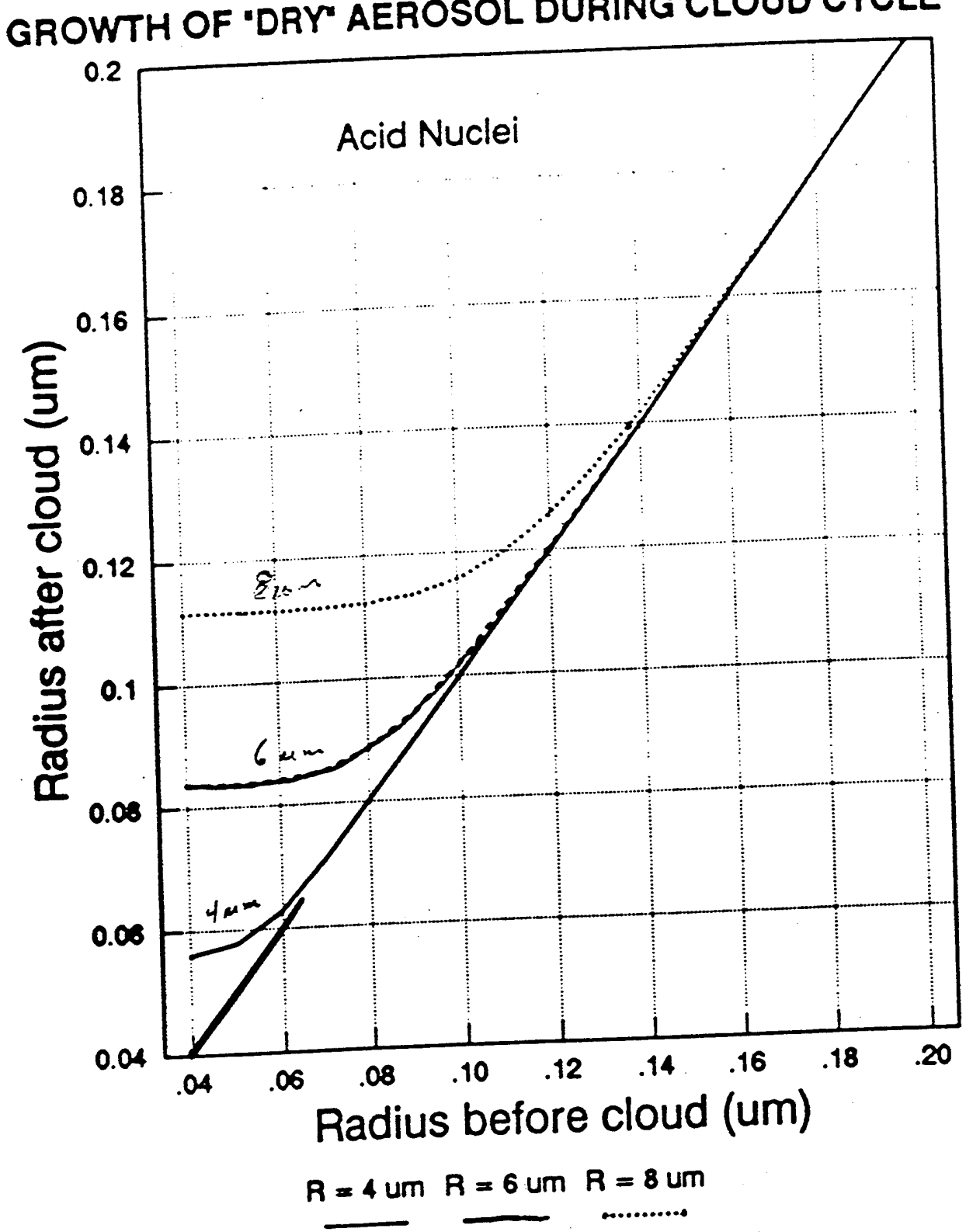

1400
1200
1000
800
600
400
200
0
z(m)
0.0
1.0
2.0
3.0
4.0
5.0
6.0
7.0
$\beta_e(z)/\beta_e(z_0)$
14 MARCH 1500Z
13 MARCH 1600Z
21 MARCH 1100Z

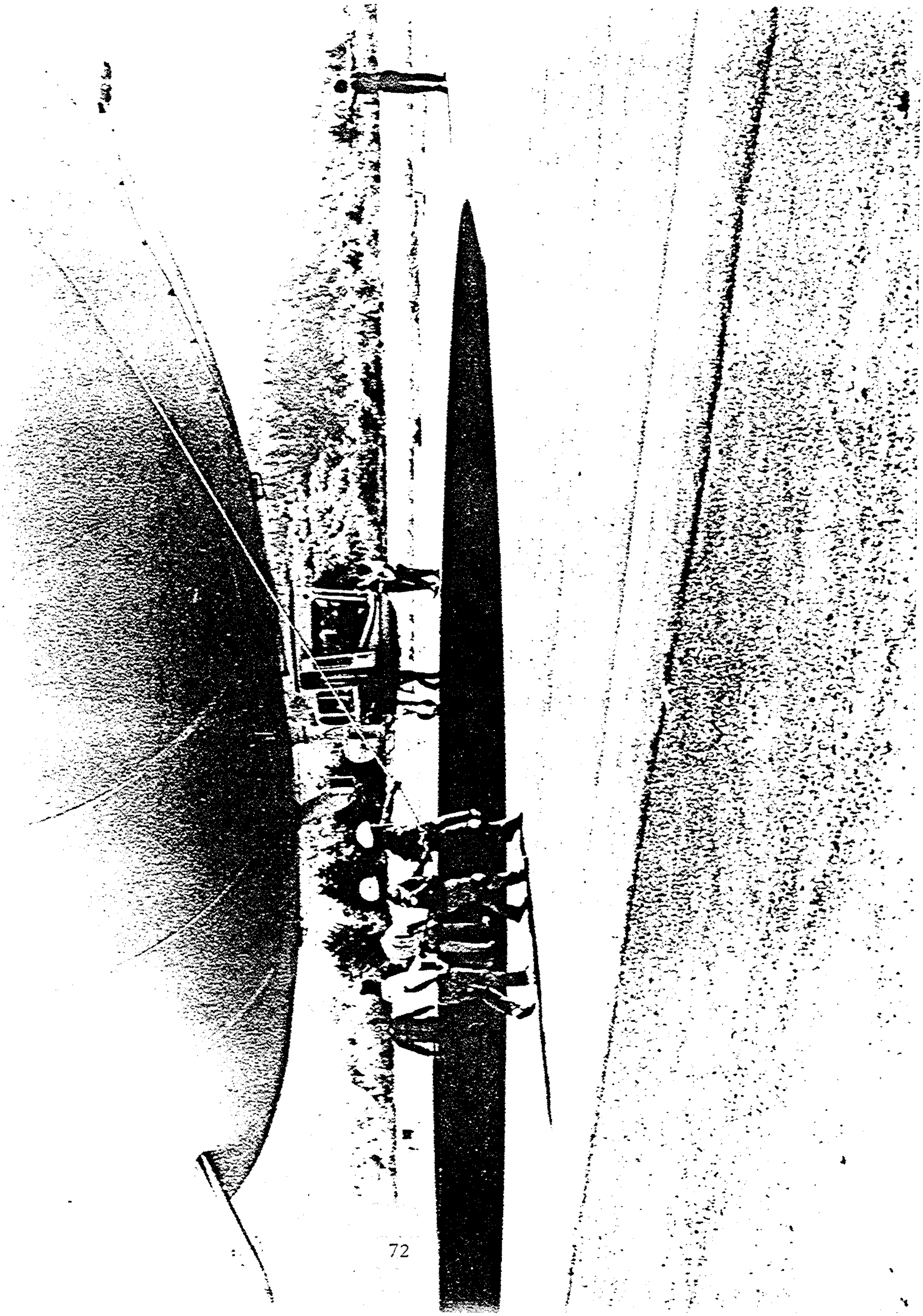

AVERAGE
HIGH AND LOW
100
90
80
70
60
50
40
30
20
10
0
% EXTINCTION AT 0.55 μm
.04-.1
.1-.3
.3-.6
.6-.9
.9-3
PARTICLE SIZE RANGE (μm RADIUS)

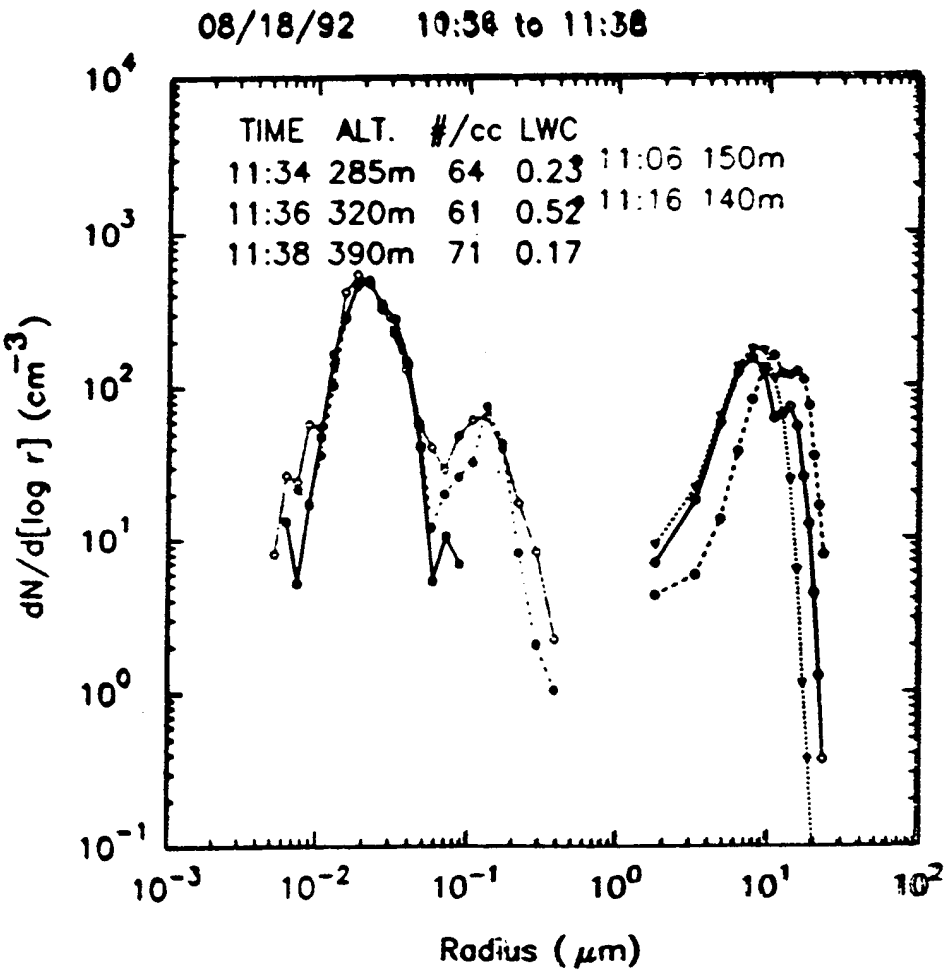

SIZE DISTRIBUTION
INITIAL
01
0.1
1.0
10.0
SIZE DISTRIBUTION
IN CLOUD
CLOUD
DROPLETS
.01
0.1
1.0
10.0
SIZE DISTRIBUTION
EVAPORATED
CLOUD
.01
0.1
1.0
10.0
RADIUS (μm)

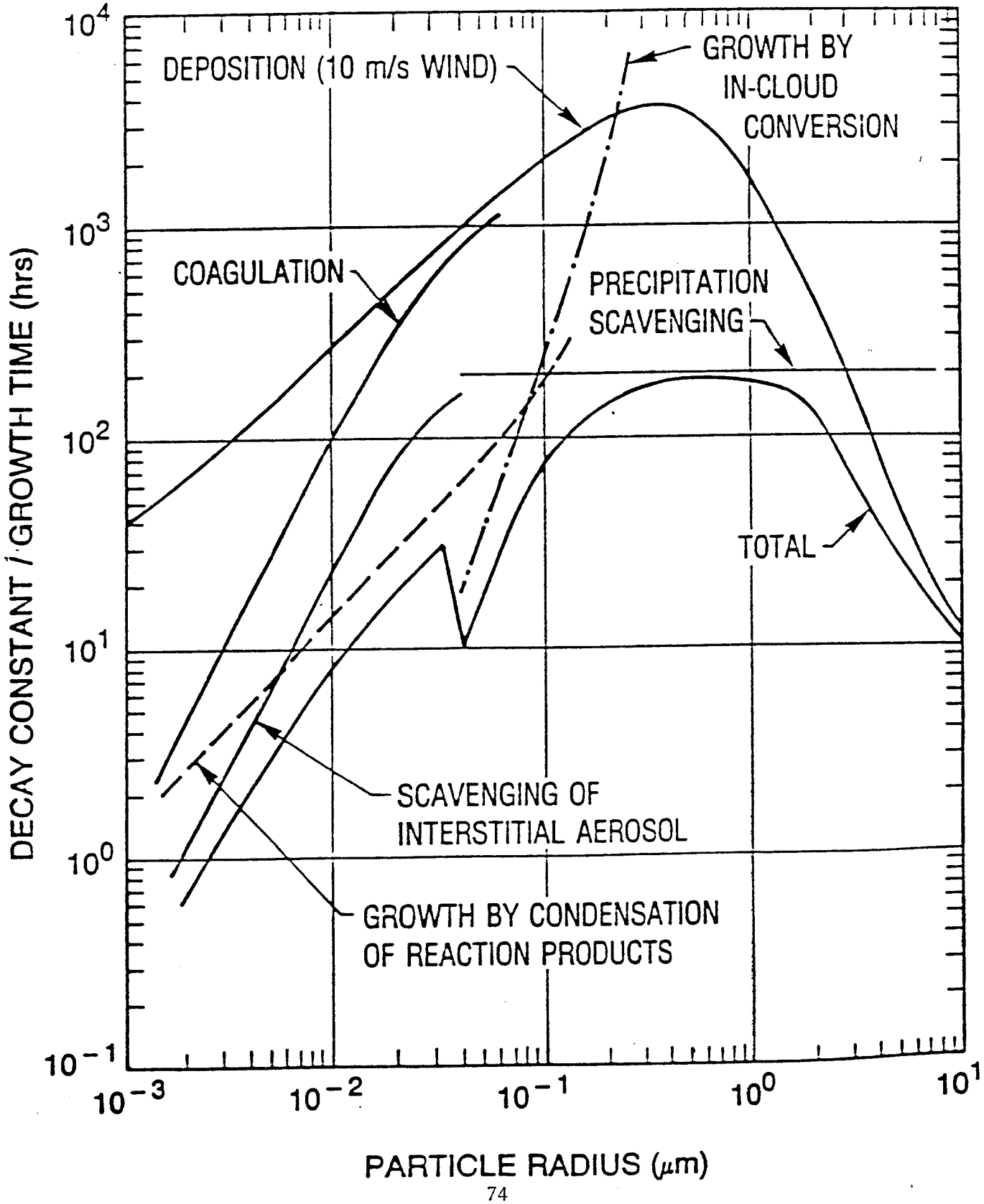

1000000
100000
10000
1000
100
10
DN/DR (NO./CUBIC CM/MICRON)
NEW PARTICLE FORMATION
CLOUD DROPLET RESIDUE
.001
.01
.1
1
RADIUS (MICRONS)


11/09/90
10^5
10^4
10^3
10^2
10^1
10^0
dN/d(log r)
.001
.01
.1
1
RADIUS (um)


GROWTH OF "DRY" AEROSOL DURING CLOUD CYCLE
Acid Nuclei
Radius after cloud (um)
0.2
0.18
0.16
0.14
0.12
0.1
0.08
0.06
0.04
.04
.06
.08
.10
.12
.14
.16
.18
.20
Radius before cloud (um)
R = 4 um
R = 6 um
R = 8 um
Ozone = 90 ppb
SO2 = 6 ppb
Time in cloud is 4 minutes




08/18/92 10:38 to 11:38
TIME ALT. #/cc LWC
11:34 285m 64 0.23
11:36 320m 61 0.52
11:38 390m 71 0.17
11:06 150m
11:16 140m
dN/d[log r] (cm^{-3})
Radius (μm)


DEPOSITION (10 m/s WIND)
GROWTH BY IN-CLOUD CONVERSION
COAGULATION
PRECIPITATION SCAVENGING
TOTAL
SCAVENGING OF INTERSTITIAL AEROSOL
GROWTH BY CONDENSATION OF REACTION PRODUCTS
DECAY CONSTANT / GROWTH TIME (hrs)
PARTICLE RADIUS (μm)
10^4
10^3
10^2
10^1
10^0
10^{-1}
10^{-3}
10^{-2}
10^{-1}
10^0
10^1

MODELING MULTICOMPONENT AEROSOL DYNAMICS IN THE MARINE BOUNDARY LAYER

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Remote Sensing Physics Branch
Code 7228

(In Collaboration with Dr. Fred Gelbard, Sandia National Labs.)

OBJECTIVE

Develop a process-oriented, one-dimensional model to describe the spatial (vertical) and temporal variations of aerosol size disribution and composition in the marine boundary layer

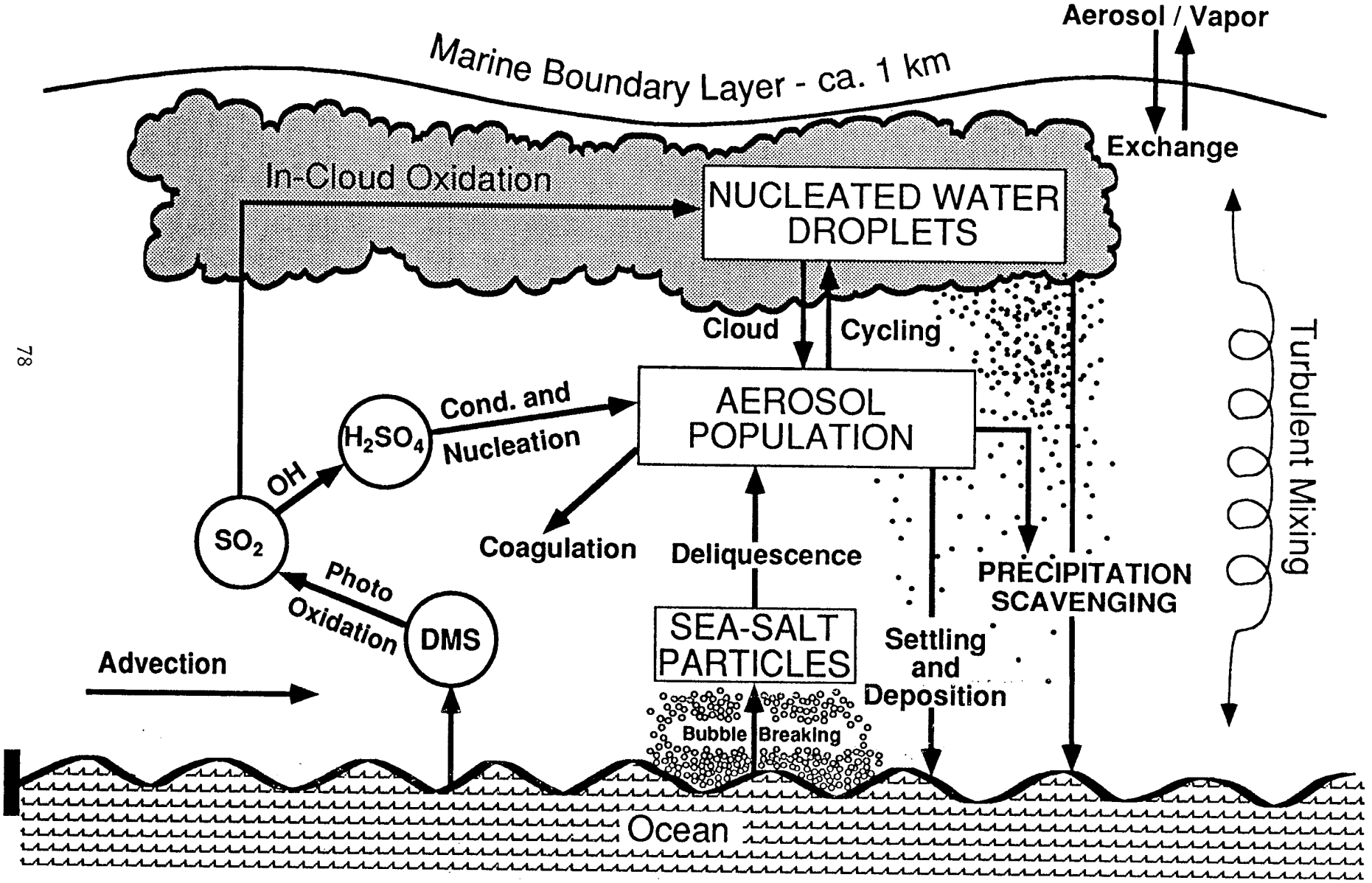
PURPOSE

- **Improve our understanding of the physical and chemical mechanisms determining the number, size distribution and composition of marine aerosols and the relative importance of these mechanisms.**

- **Aid in the interpretation of marine aerosol size distribution measurements. Simulate the transformation dynamics of aerosols in air masses advecting off the east coast of the U.S. (i.e., change in an air column moving with the wind).**

- **Describe and/or predict the temporal and spatial variation of aerosol extinction coefficient, and thus slant path optical thickness in the MBL, to improve evaluation and prediction of E-O systems performance.**

TRANSFORMATION DYNAMICS OF MARINE AEROSOLS



Governing Mechanisms

- Turbulent Mixing - One-Dimensional K-Theory
- Gravitational Settling and Deposition
- Condensation - Water Vapor (RH Change)
  - Low-Volatility Vapors (e.g. H_2SO_4)
- Coagulation - Brownian, Gravitational and Turbulent
- Sources - Nucleation and Sea-Salt Flux
- Cloud Cycling - Liquid-Phase Conversion of SO_2 to Sulfate
- Exchange between MBL and Free Troposphere

DYNAMIC AEROSOL MODEL

$$\frac{\partial n(r,z,t)}{\partial t} = \frac{\partial}{\partial z}\left[K(z,t)\,\frac{\partial n}{\partial z}\right] - v_T(r)\,\frac{\partial n(r,z,t)}{\partial z} + S(r,z,t) - L(r,z,t) + \underbrace{\frac{\partial}{\partial r}\left[n(r)\,\frac{dr}{dt}\right] + \left[\frac{\partial n}{\partial t}\right]_{COAG} + \left[\frac{\partial n}{\partial t}\right]_{CLOUD}}$$

TURBULENT MIXING

SOURCES

- SEA SALT
- NUCLEATION

SINKS

- SURFACE DEPOSITION
- PRECIPITATION SCAVENGING

TRANSFORMATIONS

- CONDENSATION
- COAGULATION
- CLOUD CYCLING

DYNAMIC AEROSOL MODEL

$$\frac{\partial n(r,z,t)}{\partial t} = \frac{\partial}{\partial z}\left[K(z,t)\,\frac{\partial n}{\partial z}\right] - v_T(r)\,\frac{\partial n(r,z,t)}{\partial z} + S(r,z,t) - L(r,z,t) + \frac{\partial}{\partial r}\left[n(r)\,\frac{dr}{dt}\right] + \left[\frac{\partial n}{\partial t}\right]_{COAG} + \left[\frac{\partial n}{\partial t}\right]_{CLOUD}$$

$$\frac{\partial \rho_{DMS}(z,t)}{\partial t} = \left[\frac{\partial \rho_{DMS}}{\partial t}\right]_{SRC} + \frac{\partial}{\partial z}\left[K(z,t)\,\frac{\partial \rho_{DMS}}{\partial z}\right] - \left[\frac{\partial \rho_{DMS}}{\partial t}\right]_{OXID}$$

$$\frac{\partial \rho_{SO_2}(z,t)}{\partial t} = \left[\frac{\partial \rho_{SO_2}}{\partial t}\right]_{SRC} + \frac{\partial}{\partial z}\left[K(z,t)\,\frac{\partial \rho_{SO_2}}{\partial z}\right] - \left[\frac{\partial \rho_{SO_2}}{\partial t}\right]_{OXID} - \left[\frac{\partial \rho_{SO_2}}{\partial t}\right]_{CLOUD} - \left[\frac{\partial \rho_{SO_2}}{\partial t}\right]_{DEP}$$

$$\frac{\partial \rho_{H_2SO_4}(z,t)}{\partial t} = \left[\frac{\partial \rho_{H_2SO_4}}{\partial t}\right]_{SRC} + \frac{\partial}{\partial z}\left[K(z,t)\,\frac{\partial \rho_{H_2SO_4}}{\partial z}\right] - \left[\frac{\partial \rho_{H_2SO_4}}{\partial t}\right]_{COND} - \left[\frac{\partial \rho_{H_2SO_4}}{\partial t}\right]_{DEP}$$

SOLUTION OF THE DYNAMIC AEROSOL EQUATION

NUMERICAL METHOD

The sectional technique (Gelbard and Seinfeld, 1979; 1980). The continuous particle size range is approximated by a finite number of size classes, or sections. The dynamic aerosol equation $\partial n(r,z,t)/\partial t$ is replaced by a set of differential equations describing the time rate of change of the mass concentration (kg/m^3) of each aerosol component, in each section and at each vertical grid level. Thus, if there are M sections, K components and N vertical grid levels, we have a set of M x K x N equations. The equations are integrated in time using a second-order Euler method with staggered time splitting. Mass and number concentrations in a section are related by a number mean value of particle mass in a section. All particles within a section are assumed to have the same composition.

INPUTS

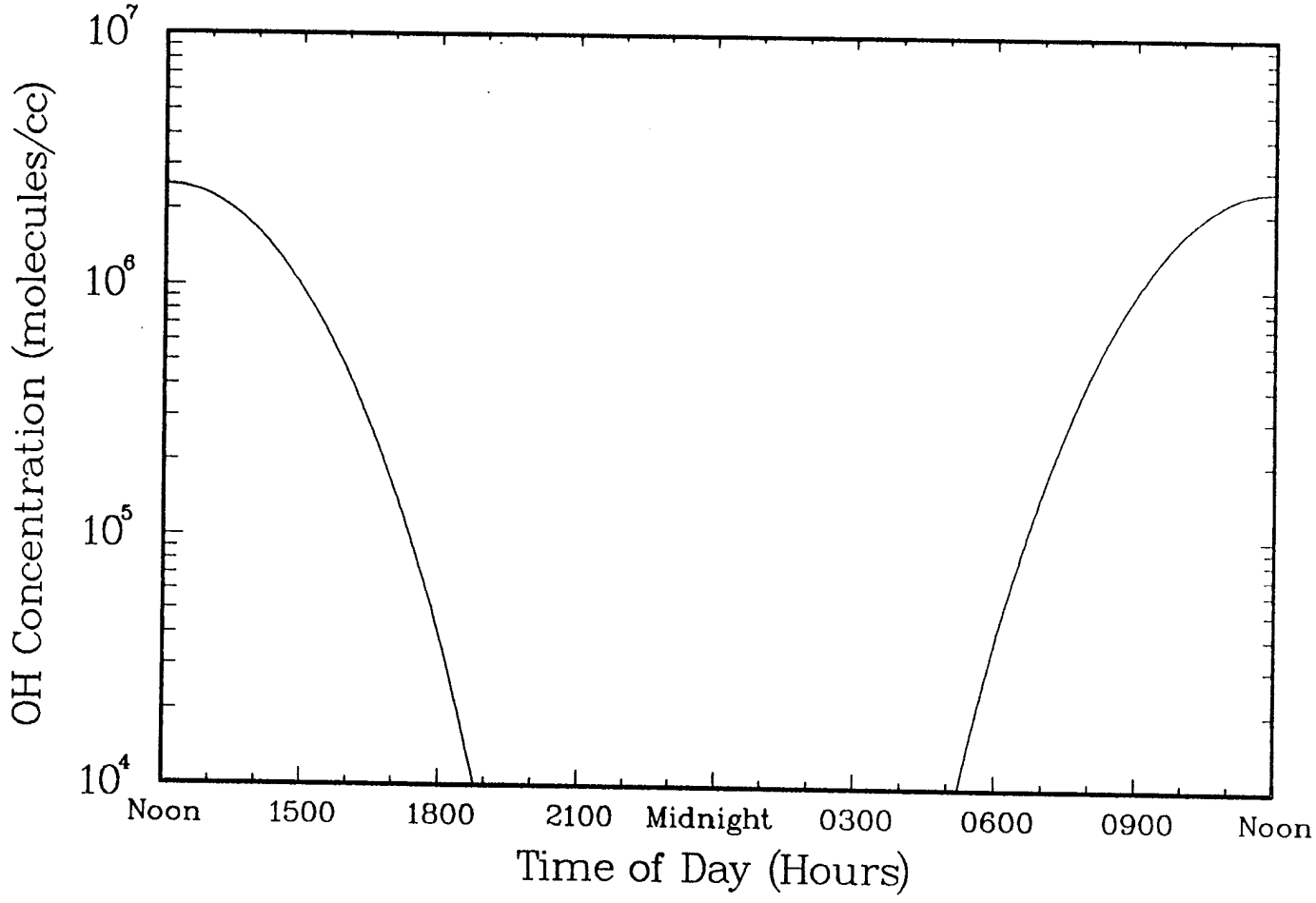
Initial mass concentrations of all components in all sections; number of sections, components and cells; section and cell boundaries; numerous parameters related to the description of the physical processes.

OUTPUTS

Aerosol mass concentrations of each component in each size section at each height (cell) at specified output times. Also, vapor concentrations at each height.

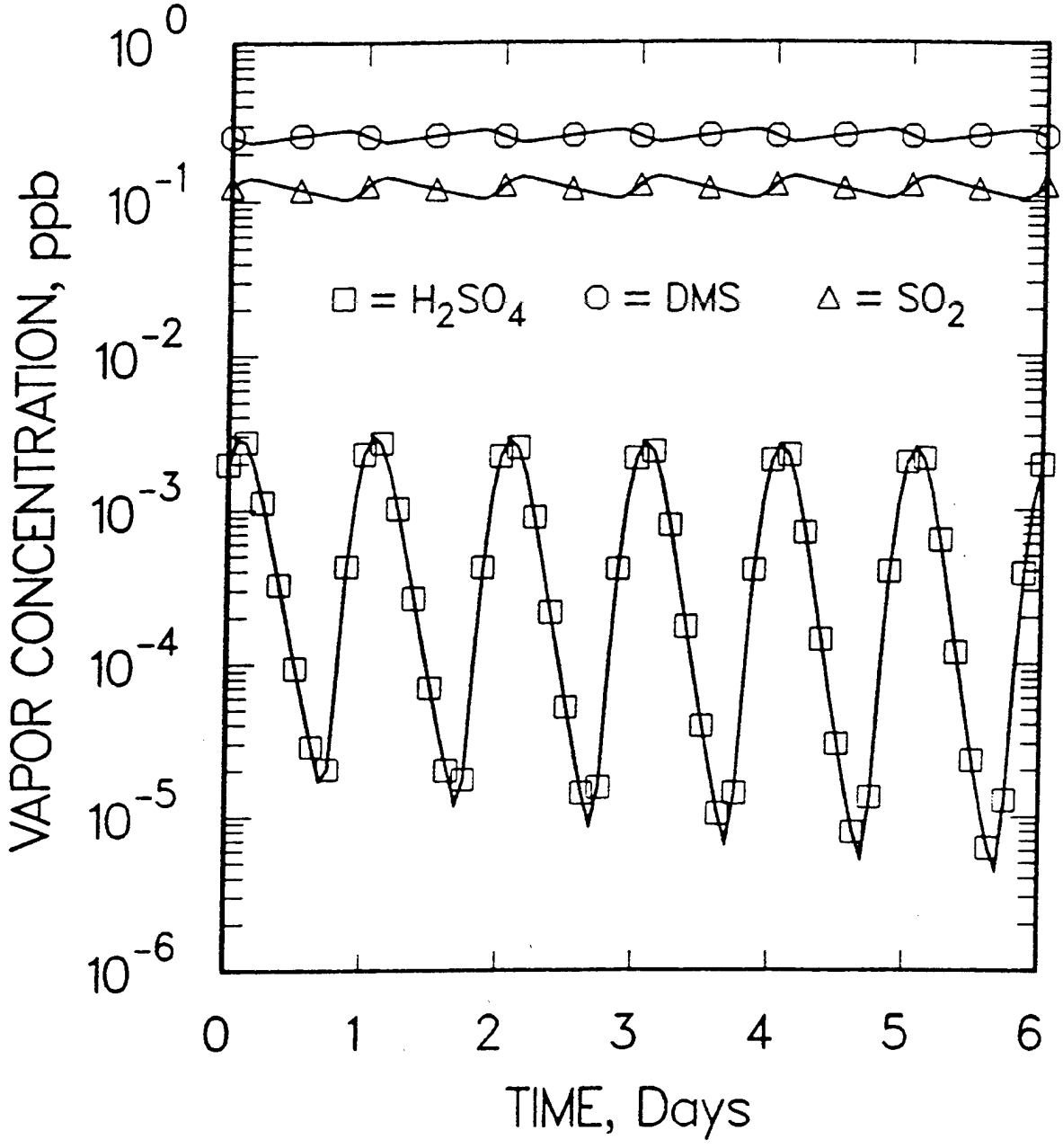
METEOROLOGICAL INPUT REQUIRED BY 1-D AEROSOL MODEL

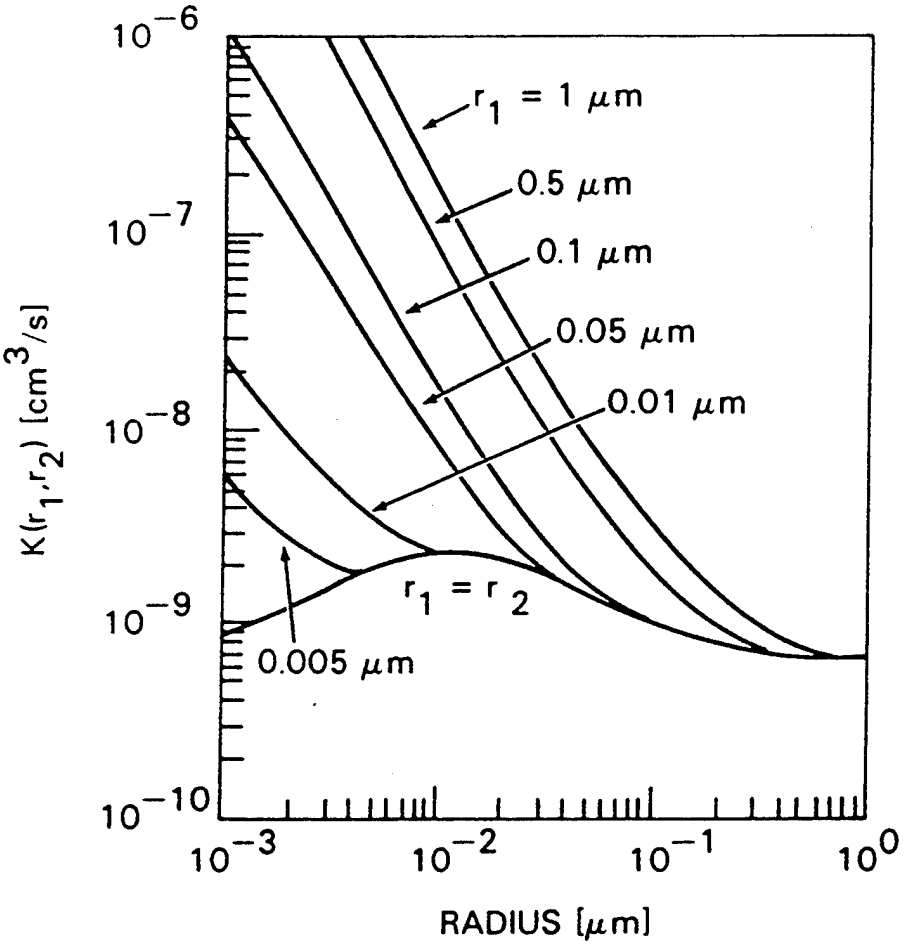
1. Vertical profile of eddy diffusivity as a function of time
2. Temperature profiles
3. Relative humidity profiles
4. Cloud layers
5. Surface winds (10 m) (for sea-salt flux)



DIURNAL VARIATION OF OH CONCENTRATIONS USED IN THE MODEL COMPUTATIONS.

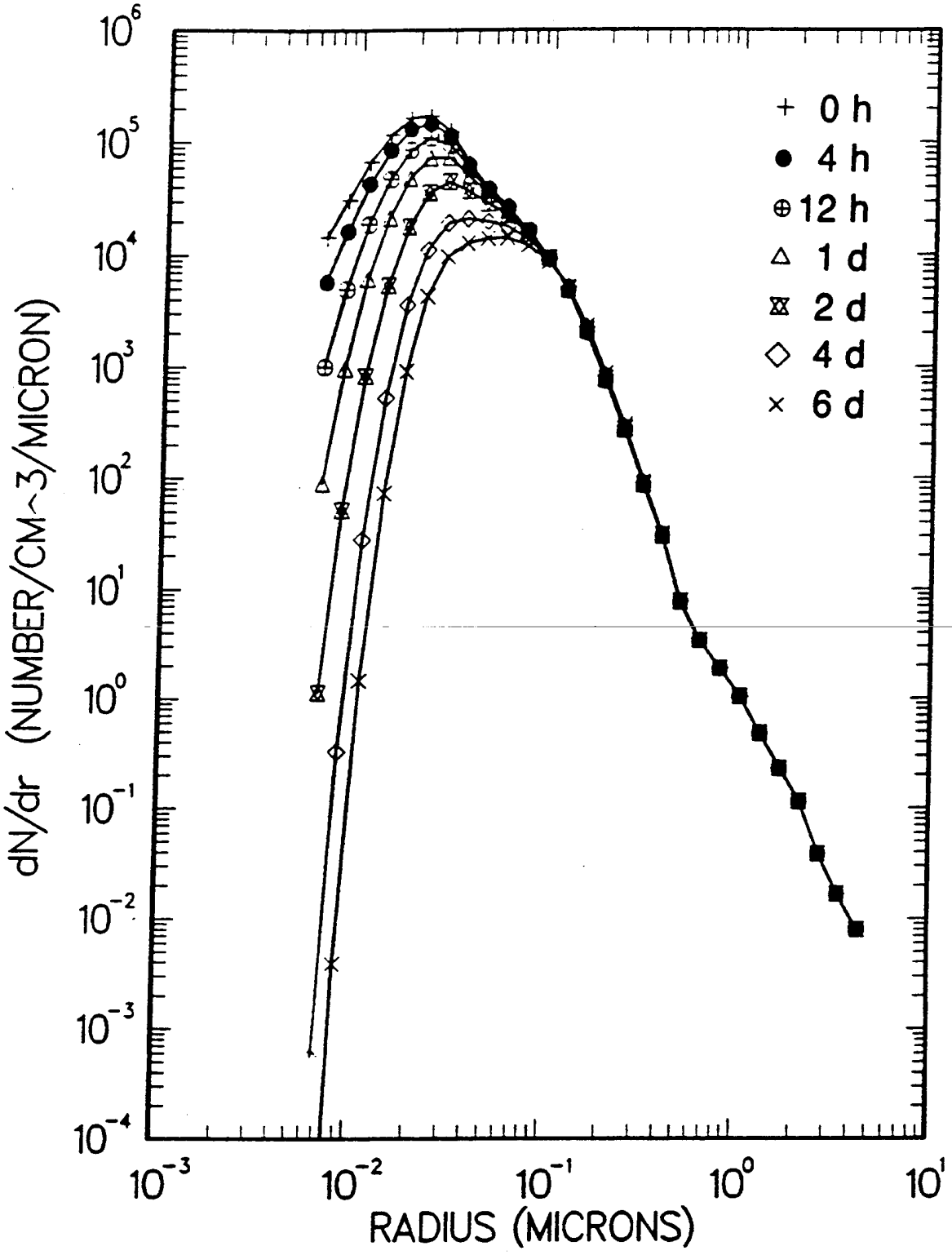
MODEL-PREDICTED EVOLUTION OF SO_2, DMS AND H_2SO_4 OVER THE REMOTE OCEAN.



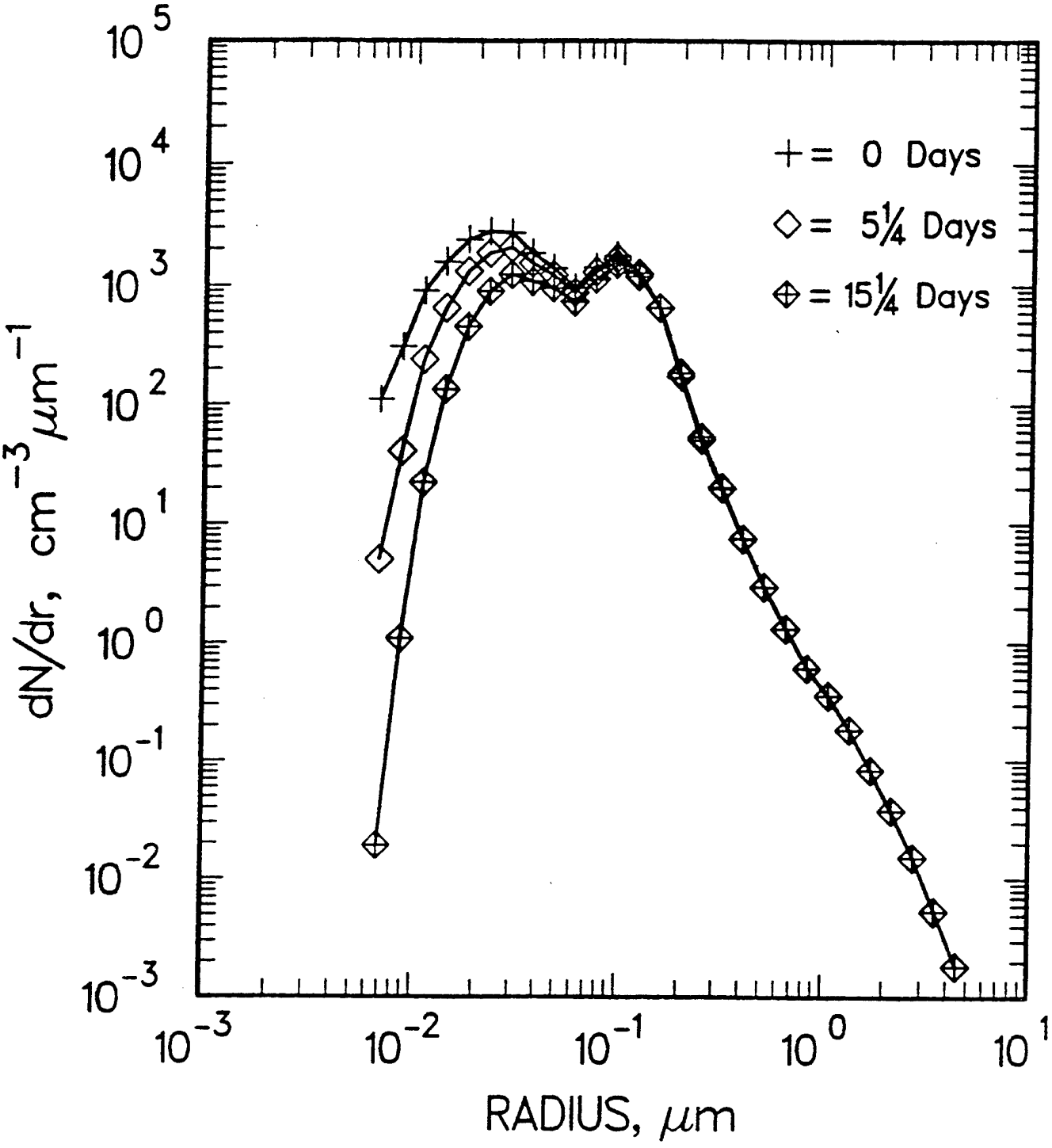


Coagulation coefficients for particles of the indicated radius with particles of smaller radii (abscissa).

MODEL-COMPUTED EFFECT OF COAGULATION AS A FUNCTION OF TIME. INITIAL DISTRIBUTION IS REPRESENTATIVE OF 4-6 HOURS OFF THE EAST COAST OF THE U. S.



MODEL SIMULATION OF THE EFFECT OF PARTICLE COAGULATION ON THE MARINE AEROSOL SIZE DISTRIBUTION. INITIAL DISTRIBUTION IS REPRESENTATIVE OF THE REMOTE ATLANTIC OCEAN.



GROWTH BY CONDENSATION OF GAS-PHASE REACTION PRODUCTS

$$\frac{dm_a}{dt} = 4\pi r D'[\rho_a(\infty) - \rho_a(r)]$$

$$\frac{dm_w}{dt} = \frac{1 - c_a}{c_a} \frac{dm_a}{dt}$$

where

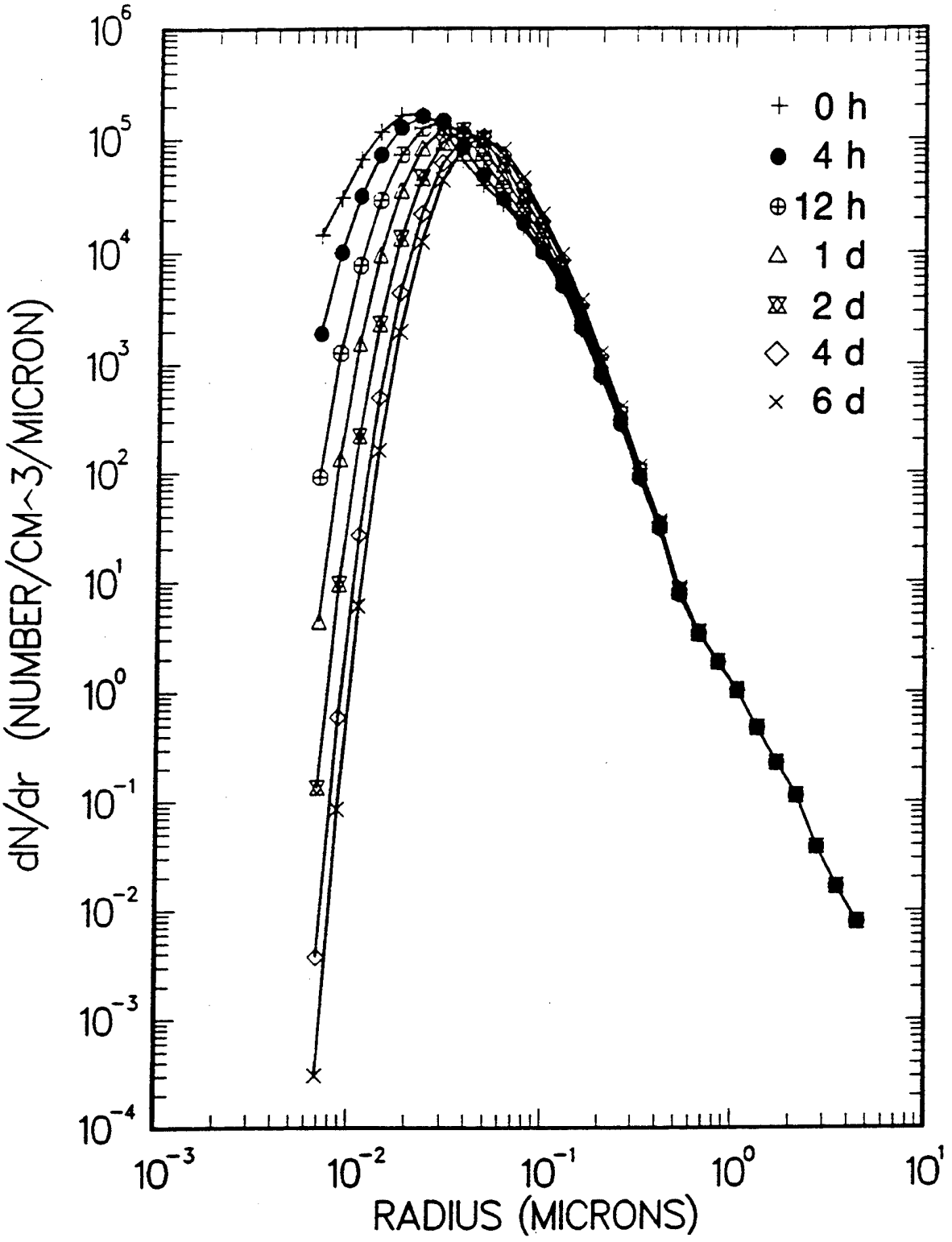
$\rho_a(\infty)$ is the environmental vapor concentration of the condensing species (e.g. H_2SO_4)
$\rho_a(r)$ is the vapor concentration at the particle surface
c_a is the weight fraction of acid in H_2O-acid solution
$D'(r)$ is the effective diffusion coefficient given by

$$D'(r) = \frac{D}{\frac{r}{r + \lambda} + \frac{4D}{r\bar{v}\alpha_c}}$$

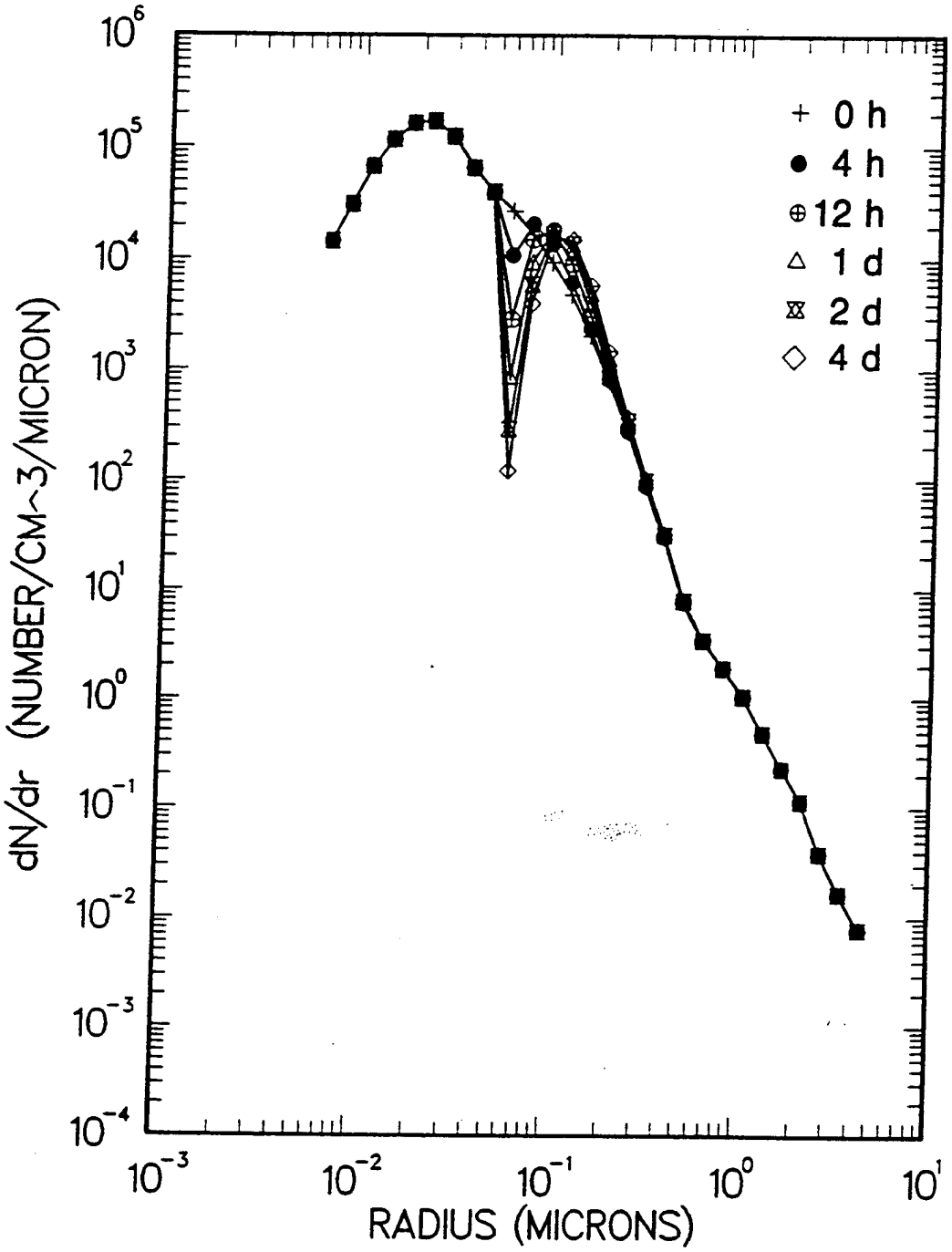
where

D is the diffusion coefficient of the condensing species
λ is its mean free path
$\bar{v}$ is the average thermal velocity of molecules of condensing species
α_c is the condensation of H_2SO_4 molecules

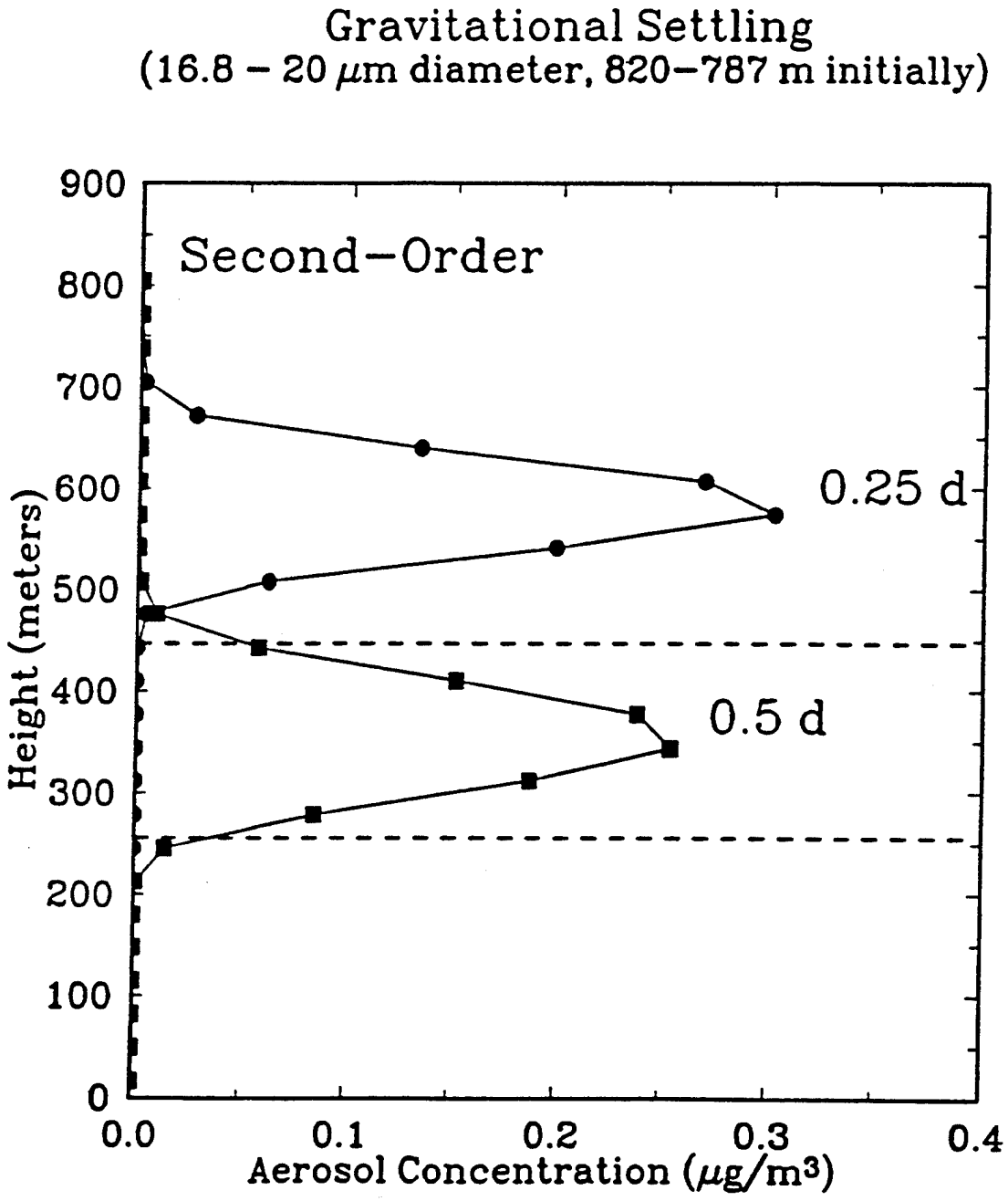
MODEL-COMPUTED EFFECT OF THE BINARY CONDENSATION OF H_2SO_4 AND WATER ON THE AEROSOL SIZE DISTRIBUTION, AS A FUNCTION OF TIME.



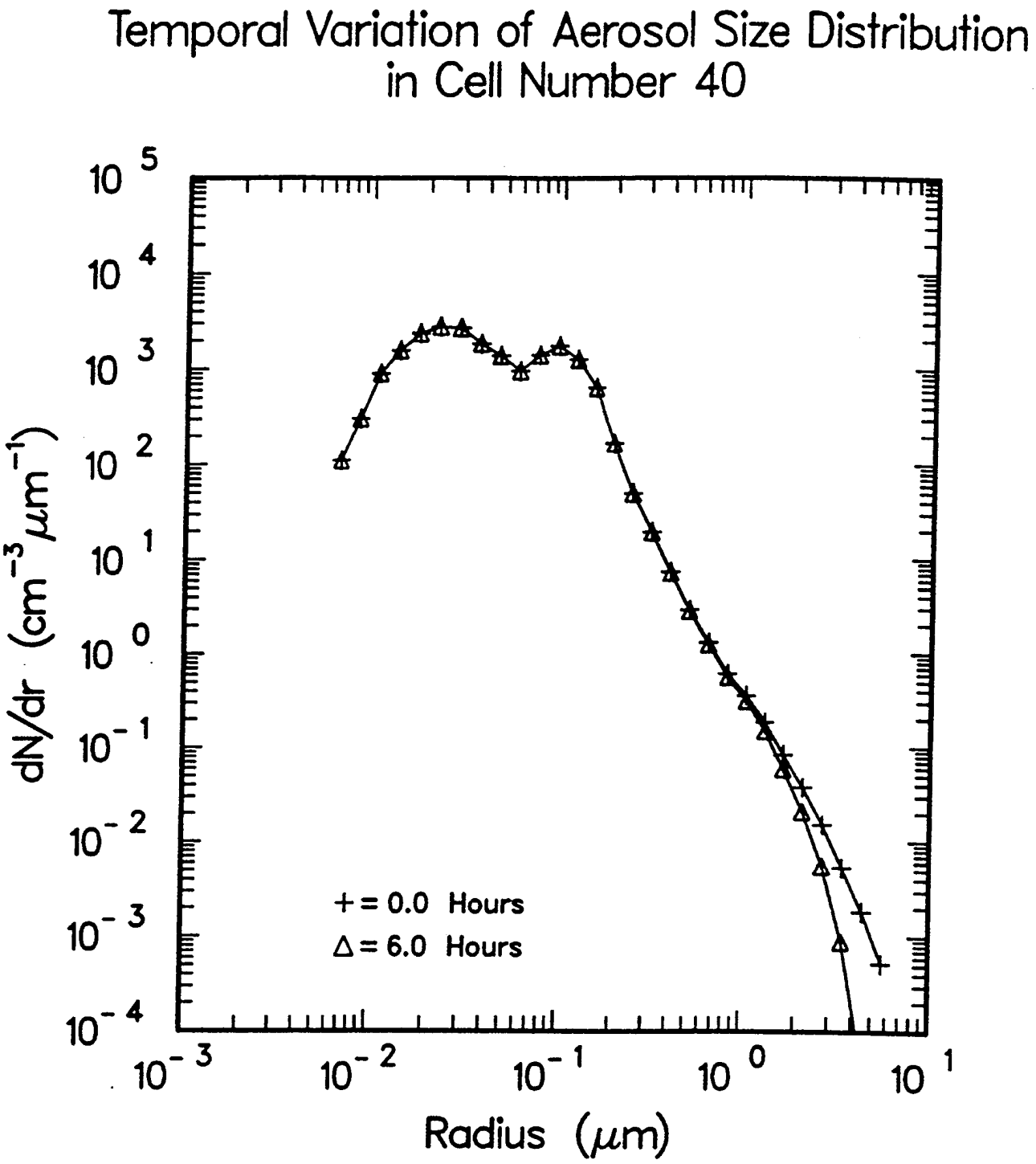
MODEL-COMPUTED EFFECT OF IN-CLOUD CONVERSION (OXIDATION) OF SO_2 TO SULFATE ON THE AEROSOL SIZE DISTRIBUTION.

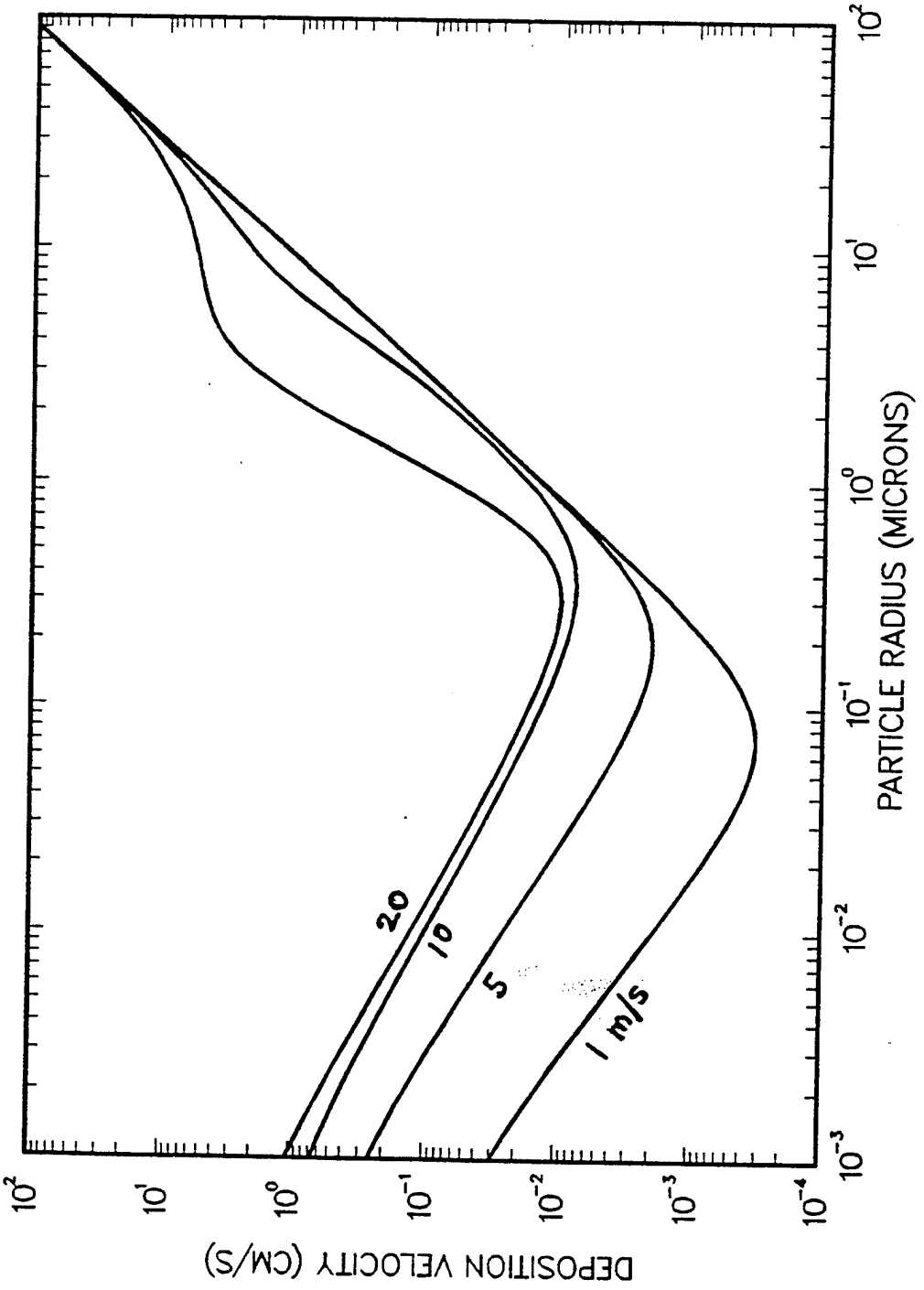


DEMONSTRATES THE ACCURACY OF THE NUMERICAL METHOD USED TO COMPUTE GRAVITATIONAL SETTLING. ANALYTICAL COMPUTATIONS SHOW THAT PARTICLES IN THE INDICATED SIZE RANGE, AND INITIALLY BETWEEN 787-820 METERS, SHOULD LIE BETWEEN THE DASHED LINES AFTER 12 HOURS



CHANGE IN THE PARTICLE SIZE DISTRIBUTION IN THE TOP 25 METERS OF THE BOUNDARY LAYER AFTER 6 HOURS, DUE TO GRAVITATIONAL SETTLING ONLY




DEPOSITION VELOCITY (CM/S)
PARTICLE RADIUS (MICRONS)
20
10
5
1 m/s

WATER UPTAKE DUE TO HYGROSCOPICITY

For particles consisting of a mixed solute, the water content as a function of relative is (neglecting particle curvature) given by (Hanel, 1976)

$$m_w = m_s \bar{\mu} \, f/ (1 - f)$$

where

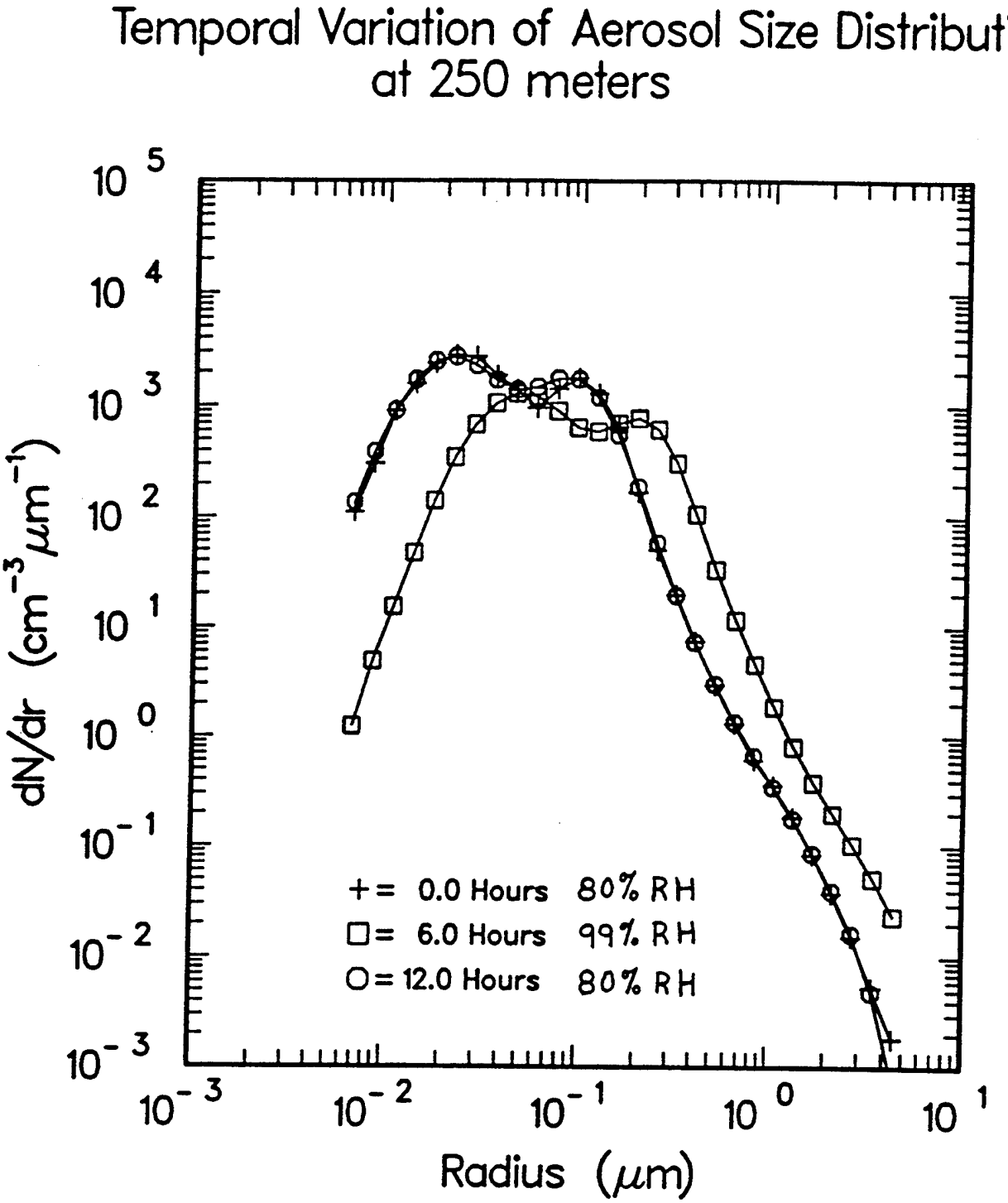
f = RH(%)/100
m_w = mass of water
m_s = mass of soluble material (i.e., sea salt + H_2SO_4 in present case)
$\bar{\mu}$ = linear mass increase coefficient for the solute mixture

$$\bar{\mu} = \Sigma \, \mu_i m_i / m_s$$

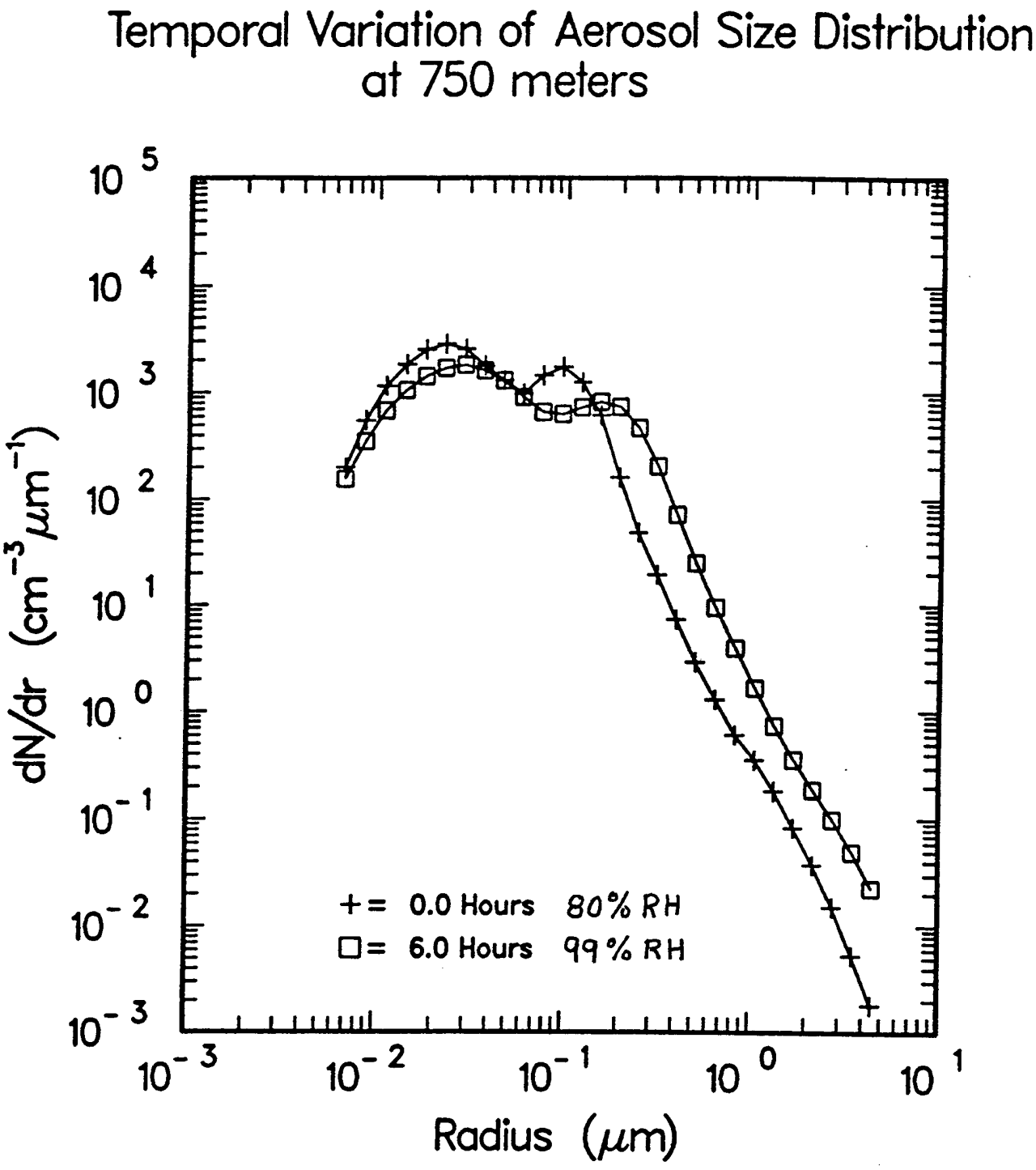
$\mu_{sea\ salt}$ is based on measurements of Keith and Arons (1954) for $a_w = f > 0.75$ and Winkler and Junge (1971) for $a_w < 0.75$

μ_{H2SO4} is determined from data on weight fraction of H_2SO_4 as a function of water activity, a_w

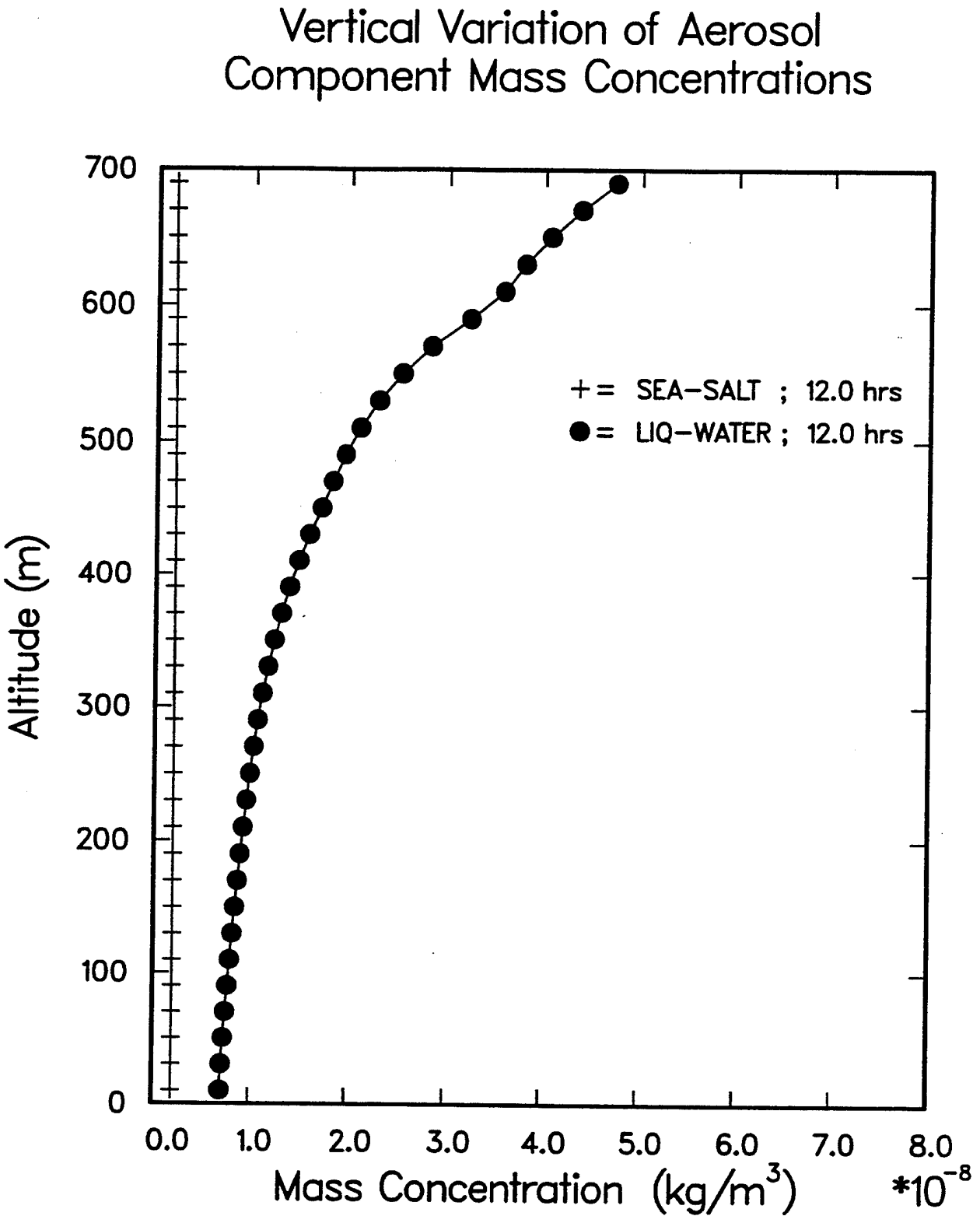
MODEL-COMPUTED CHANGE IN THE SIZE DISTRIBUTION DUE TO CHANGES IN RELATIVE HUMIDITY, SHOWING EXCELLENT REVERSIBILITY (i.e., THE INITIAL SIZE DISTRIBUTION IS RECOVERED WHEN THE RELATIVE HUMIDITY RETURNS TO ITS INITIAL VALUE



MODEL-COMPUTED CHANGE IN THE SIZE DISTRIBUTION DUE TO CHANGES IN RELATIVE HUMIDITY, WITH THE EFFECT OF PARTICLE CURVATURE INCLUDED



RELATIVE HUMIDITY VARIES FROM 80% AT THE SURFACE TO 98% AT 700 M.



SEA-SALT FLUX FORMULATIONS

- Monahan et al. (1986). Based on combining field observations of the dependence of whitecap coverage upon prevailing wind speed with estimates of aerosol production from laboratory-simulated whitecaps. The size-resolved sea-salt flux formulation consists of production terms due to bubble bursting and spume formation. The authors state that the spume production estimate has many shortcomings and its use is not recommended. Size range of applicability: $r > 0.5\mu m$.

- Smith et al. (1993). Used measurements of particle size spectra taken in the outer Hebrides at a height of about 10 m and a mean relative humidity of about 80%. Derived sea-salt flux as a function of wind speed and particle size assuming that the observed particle spectrum is an equilibrium distribution, with the loss of particles by turbulent deposition and gravitational settling balanced by sea-salt production. Size range of applicability: $r > 1\mu m$.

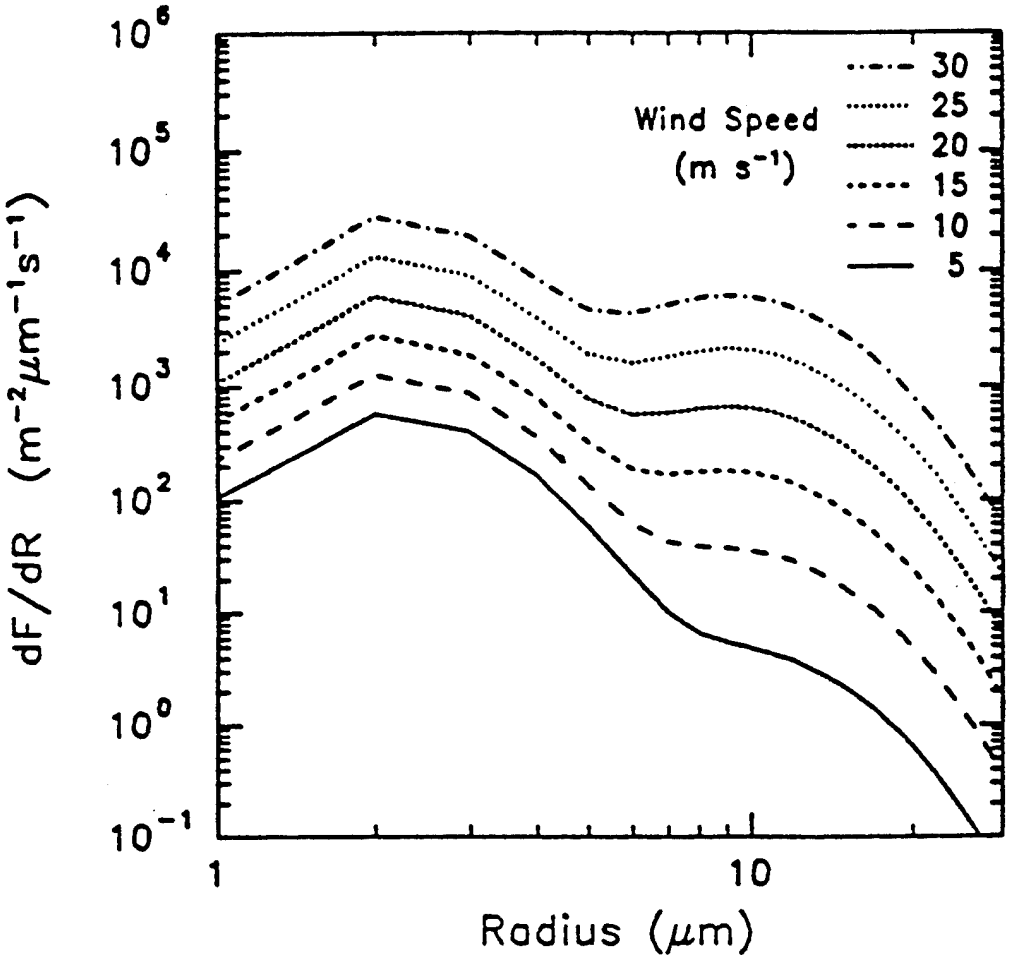


Figure 6. **Variation of the model particle fluxes with wind speed and particle size.** (according to Smith et al., 1993)

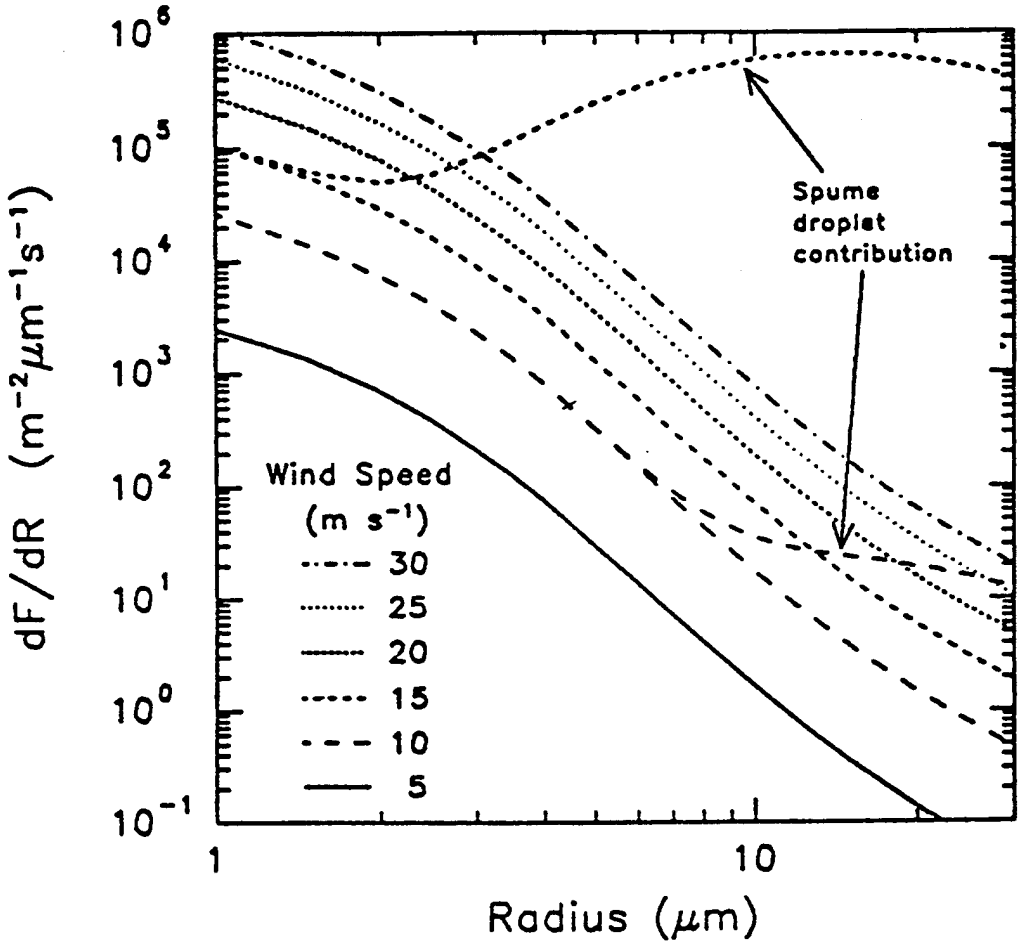
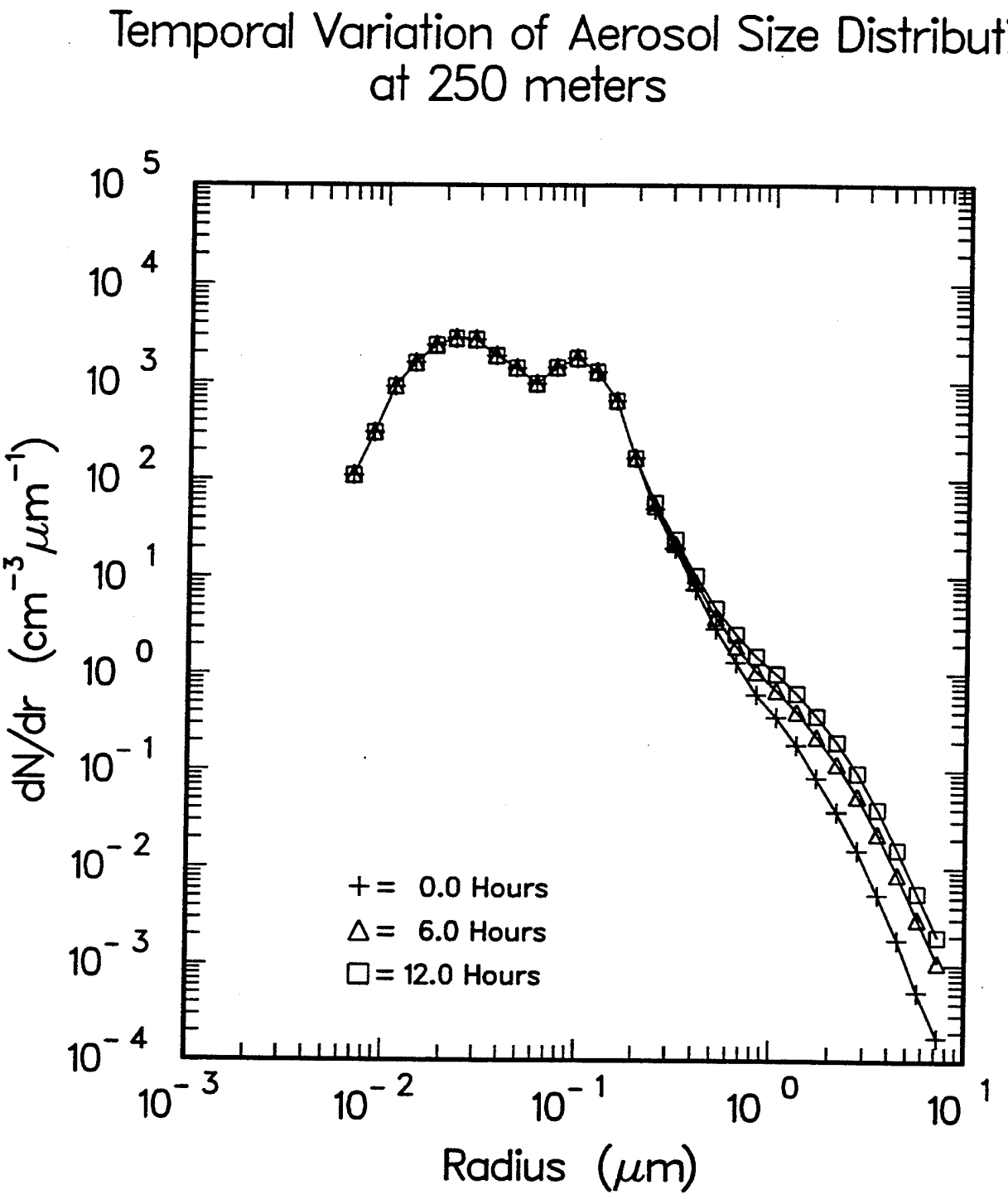
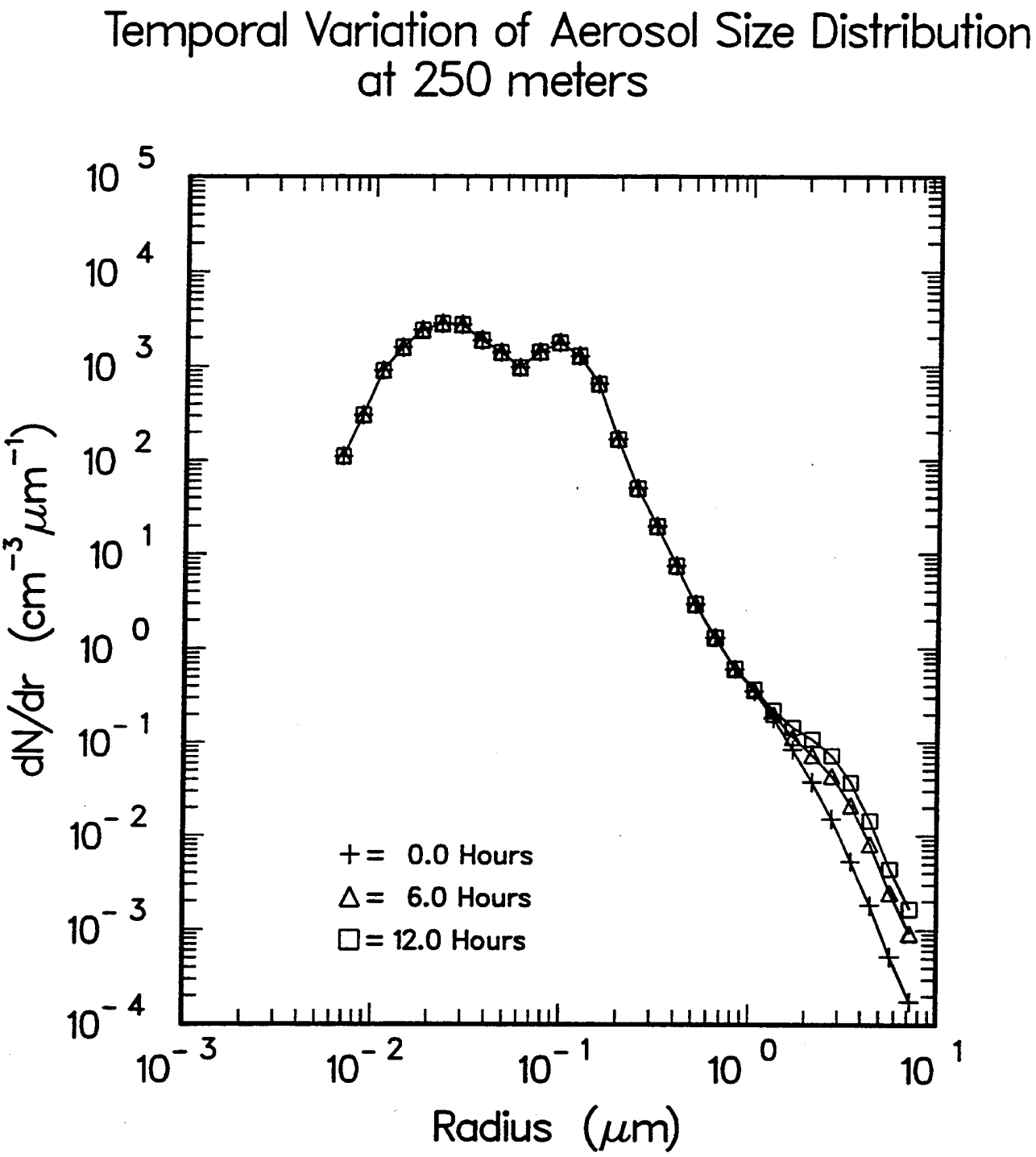


Figure 7. Whitecap and spume-droplet fluxes from Monahan *et al.* (1986).

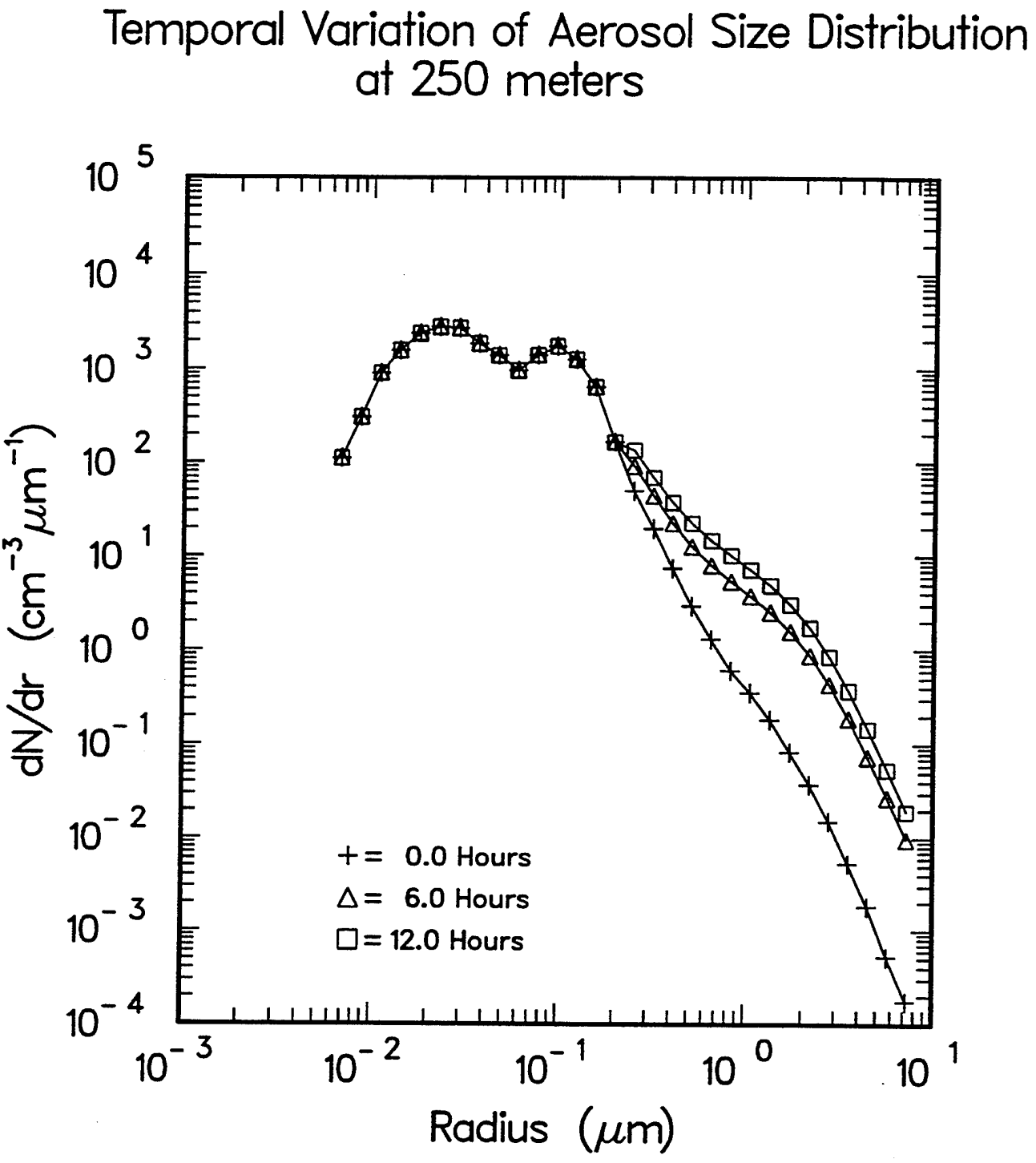
EFFECT OF SEA-SALT FLUX ON THE PARTICLE SIZE DISTRIBUTION. FLUX FORMULATION OF MONAHAN ET AL. (1986) IS ASSUMED. MBL IS 500 METERS THICK (ONE LAYER). WIND SPEED IS 7 M/SEC.



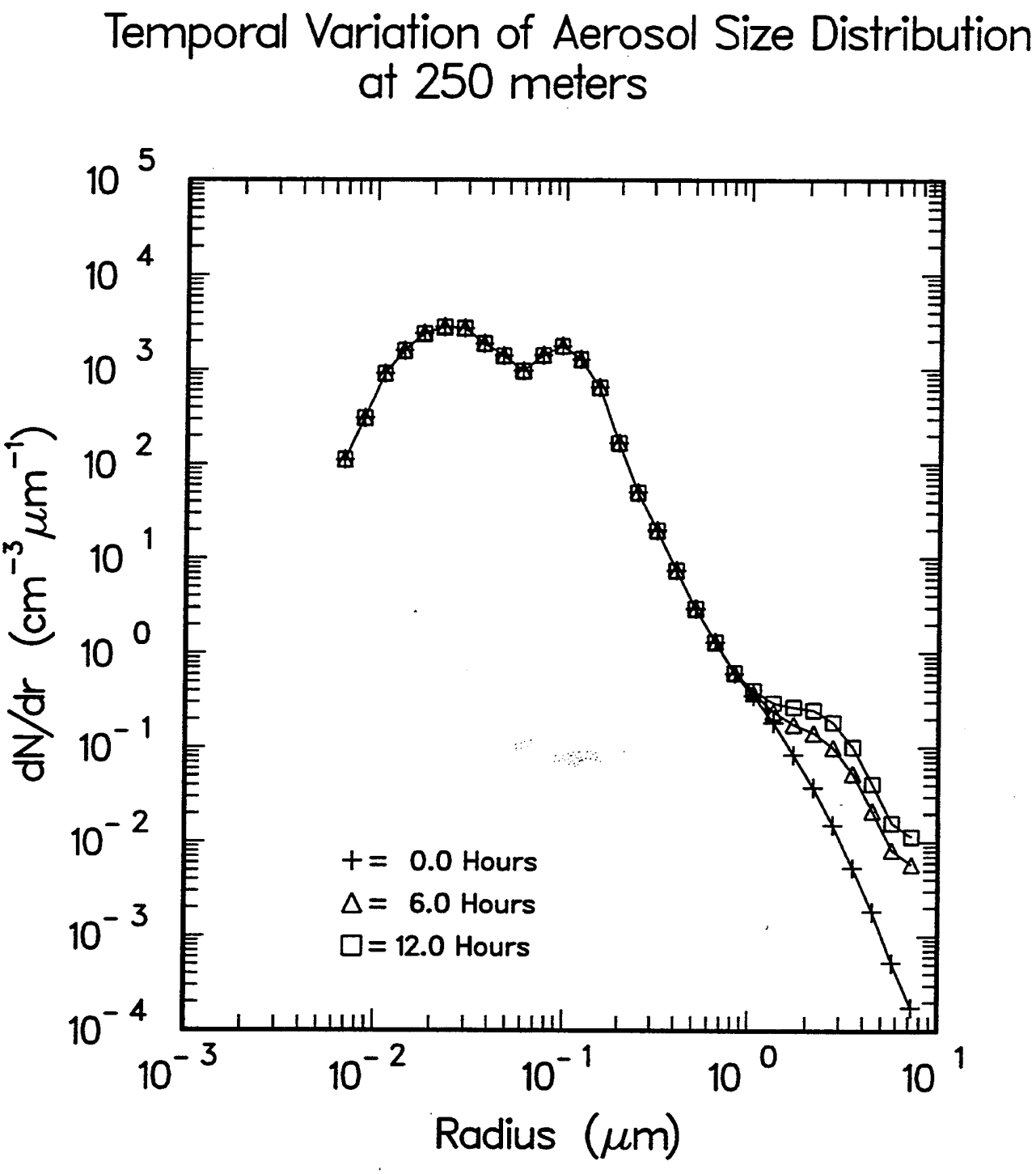
EFFECT OF SEA-SALT FLUX ON THE PARTICLE SIZE DISTRIBUTION. FLUX FORMULATION OF SMITH ET AL. (1993) IS ASSUMED. MBL IS 500 METERS THICK (ONE LAYER). WIND SPEED IS 7 M/SEC.



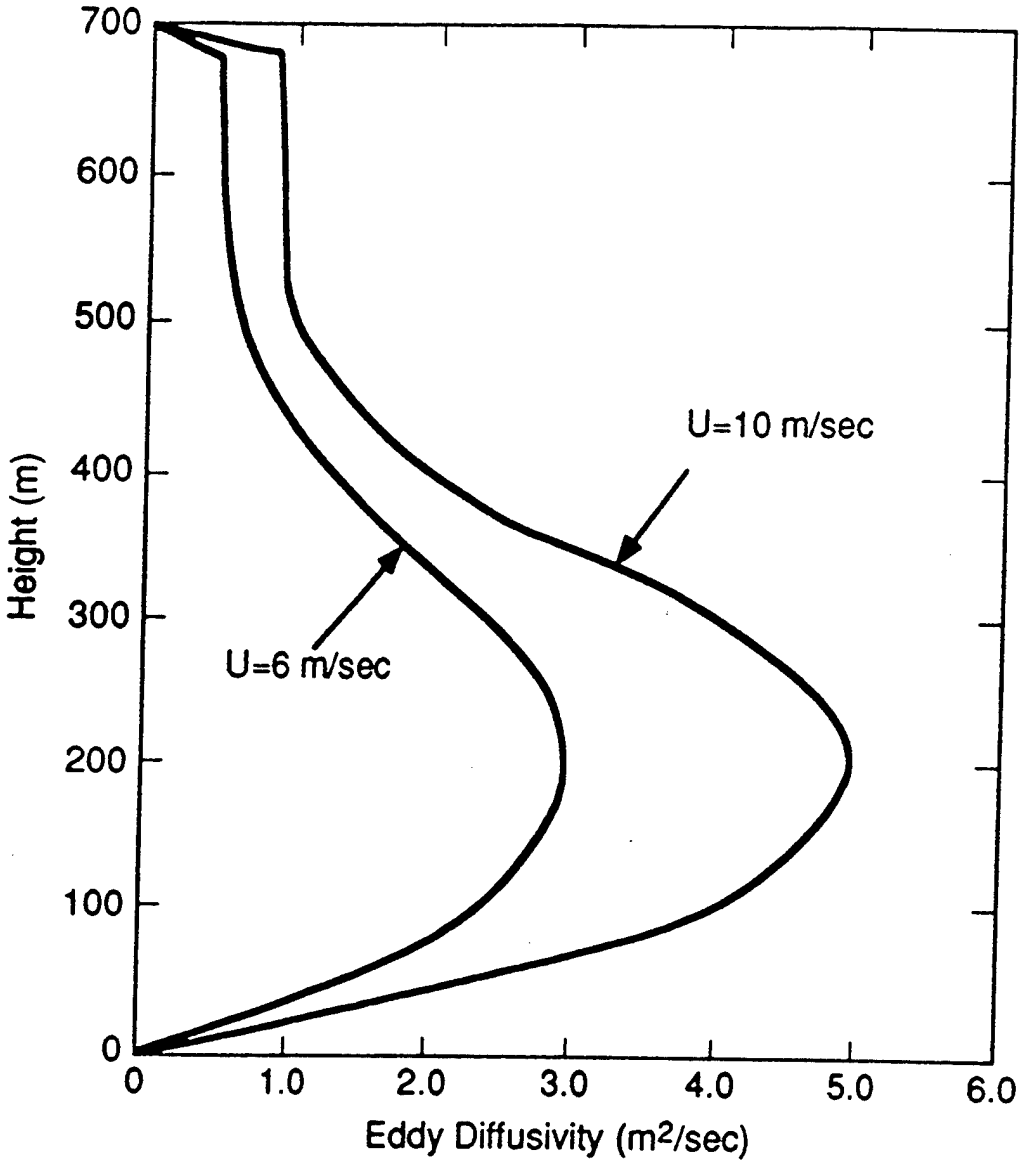
EFFECT OF SEA-SALT FLUX ON THE PARTICLE SIZE DISTRIBUTION. FLUX FORMULATION OF MONAHAN ET AL. (1986) IS ASSUMED. MBL IS 500 METERS THICK (ONE LAYER). WIND SPEED IS 14 M/SEC.



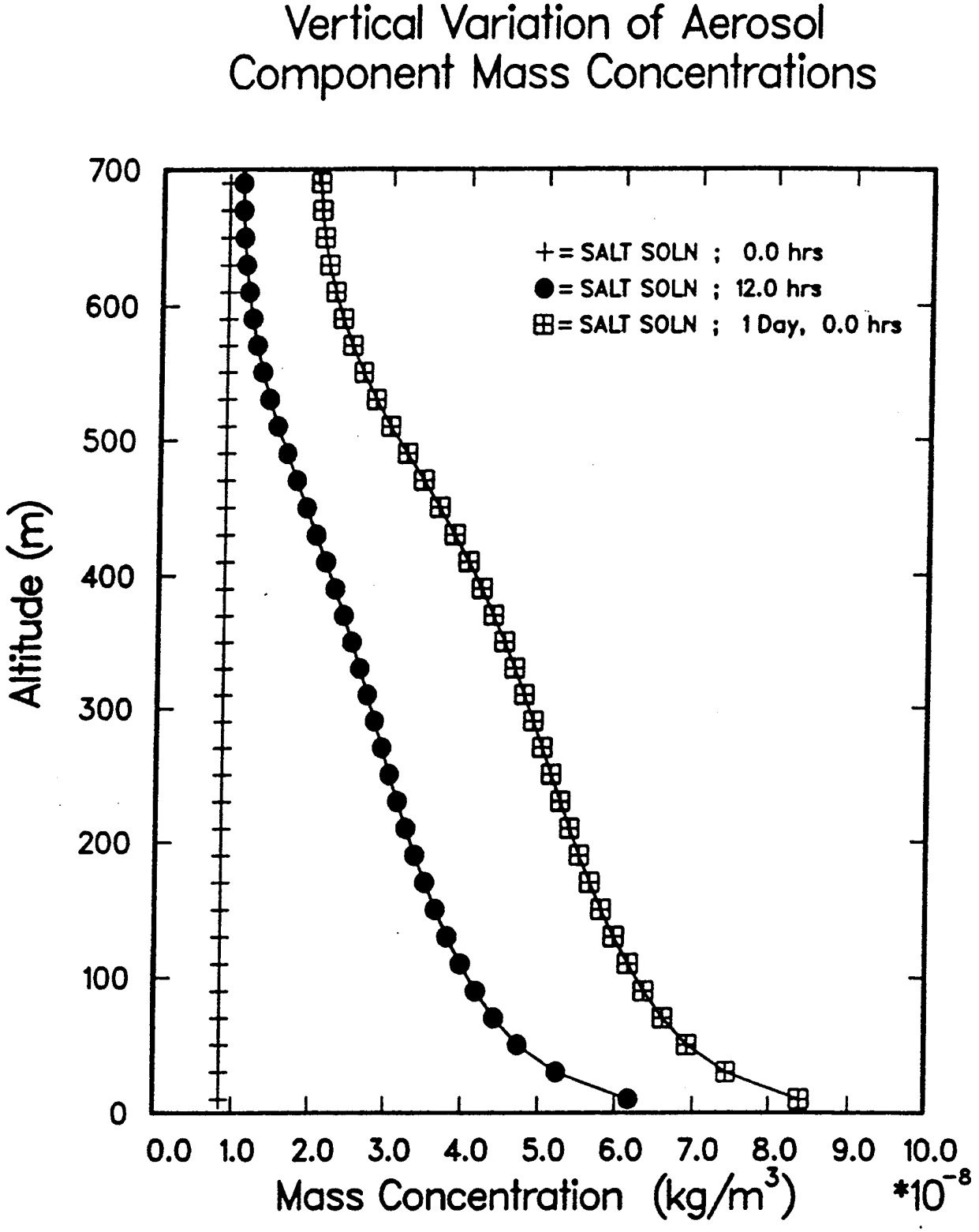
EFFECT OF SEA-SALT FLUX ON THE PARTICLE SIZE DISTRIBUTION. FLUX FORMULATION OF SMITH ET AL. (1993) IS ASSUMED. MBL IS 500 METERS THICK (ONE LAYER). WIND SPEED IS 14 M/SEC.



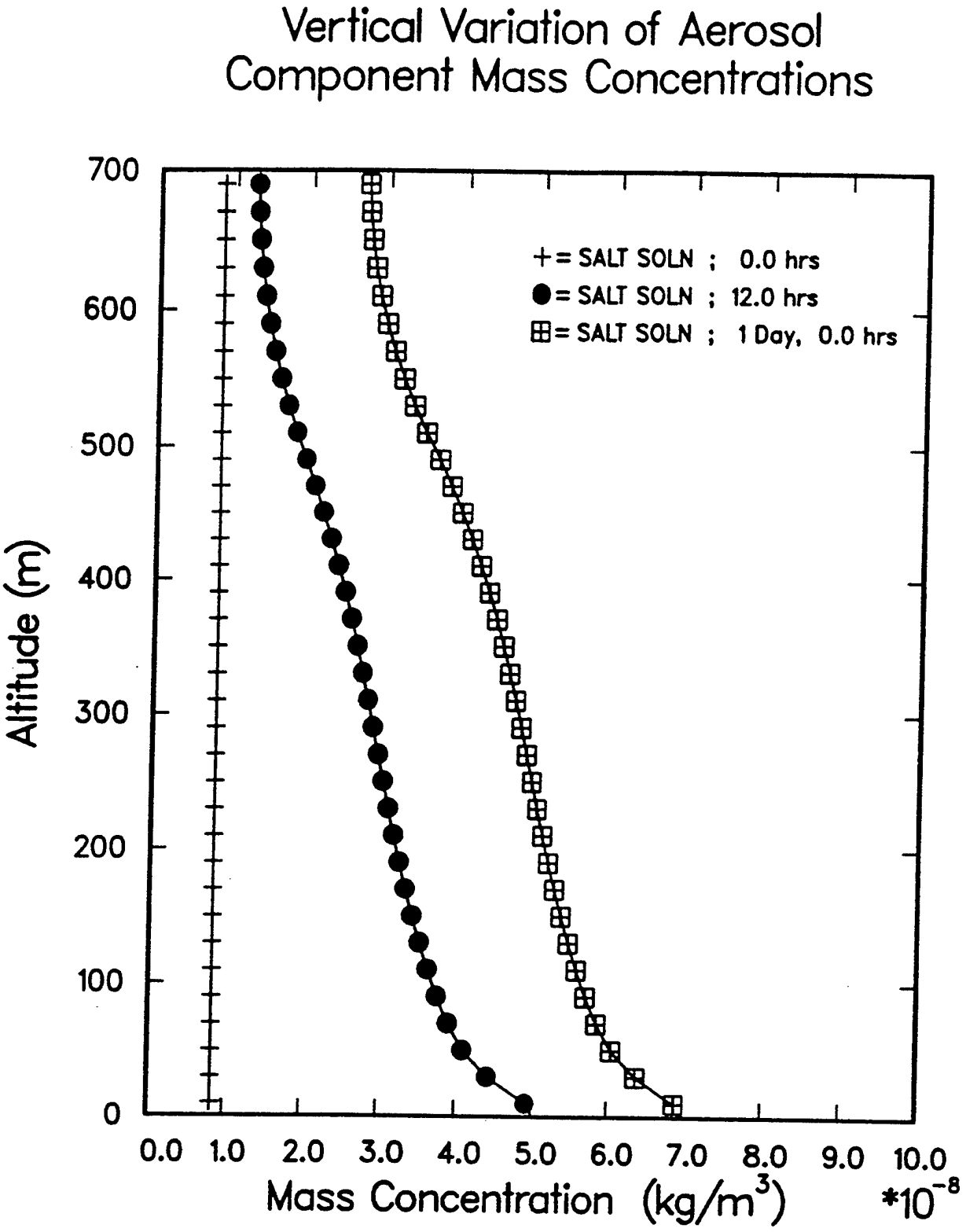
PROFILES OF EDDY DIFFUSIVITY USED FOR THE PURPOSE OF SIMULATING THE EFFECT OF SEA-SALT FLUX AND TURBULENT MIXING



VERTICAL VARIATION OF THE MASS CONCENTRATION OF SALT SOLUTION (SEA-SALT AND ASSOCIATED WATER) DUE TO FLUX FROM THE OCEAN AND TURBULENT MIXING, AS A FUNCTION OF TIME. MAXIMUM EDDY DIFFUSIVITY IS 3.0 M^2/SEC.



VERTICAL VARIATION OF THE MASS CONCENTRATION OF SALT SOLUTION (SEA-SALT AND ASSOCIATED WATER) DUE TO FLUX FROM THE OCEAN AND TURBULENT MIXING, AS A FUNCTION OF TIME. MAXIMUM EDDY DIFFUSIVITY IS 5.0 M^2/SEC.



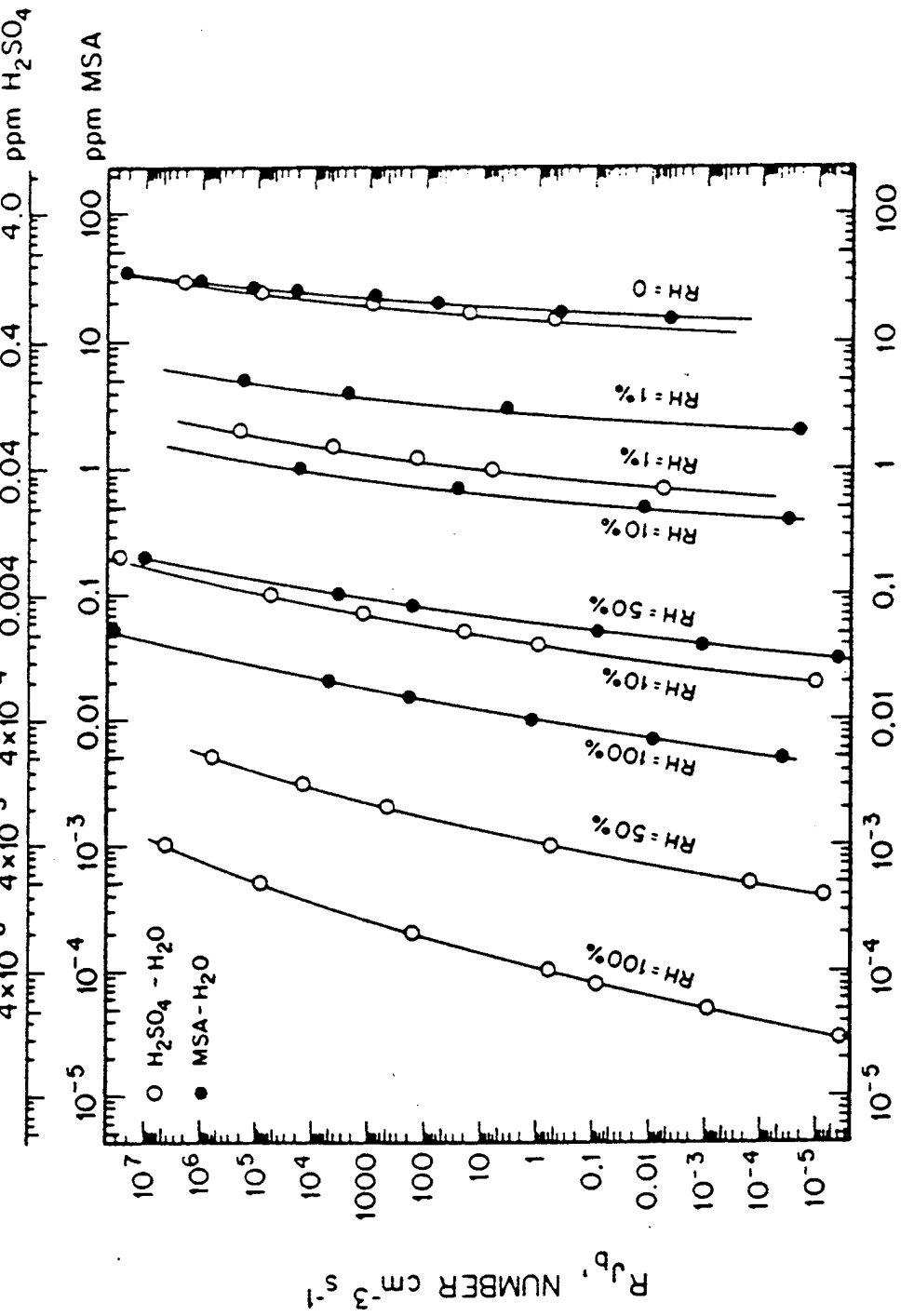
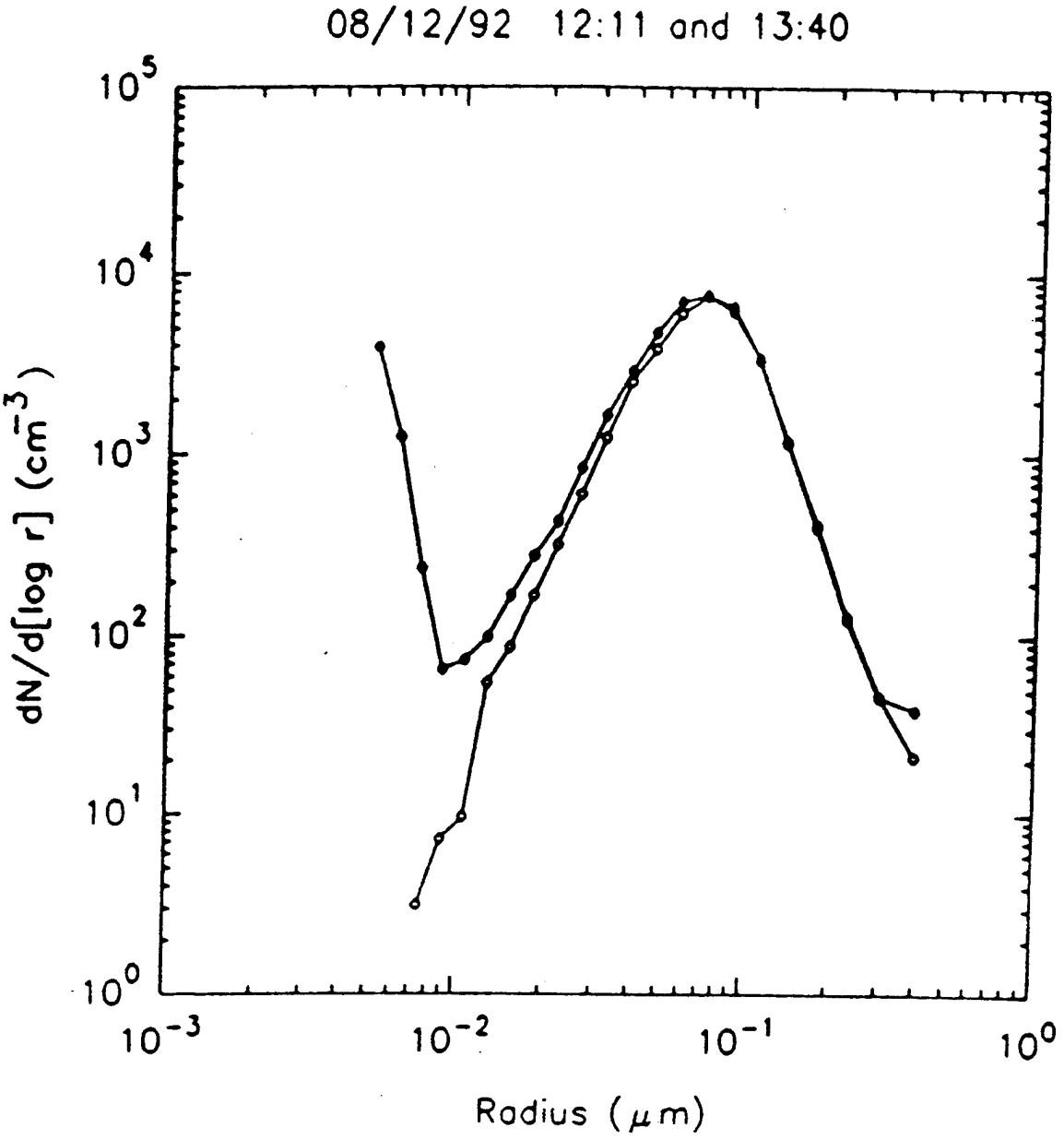
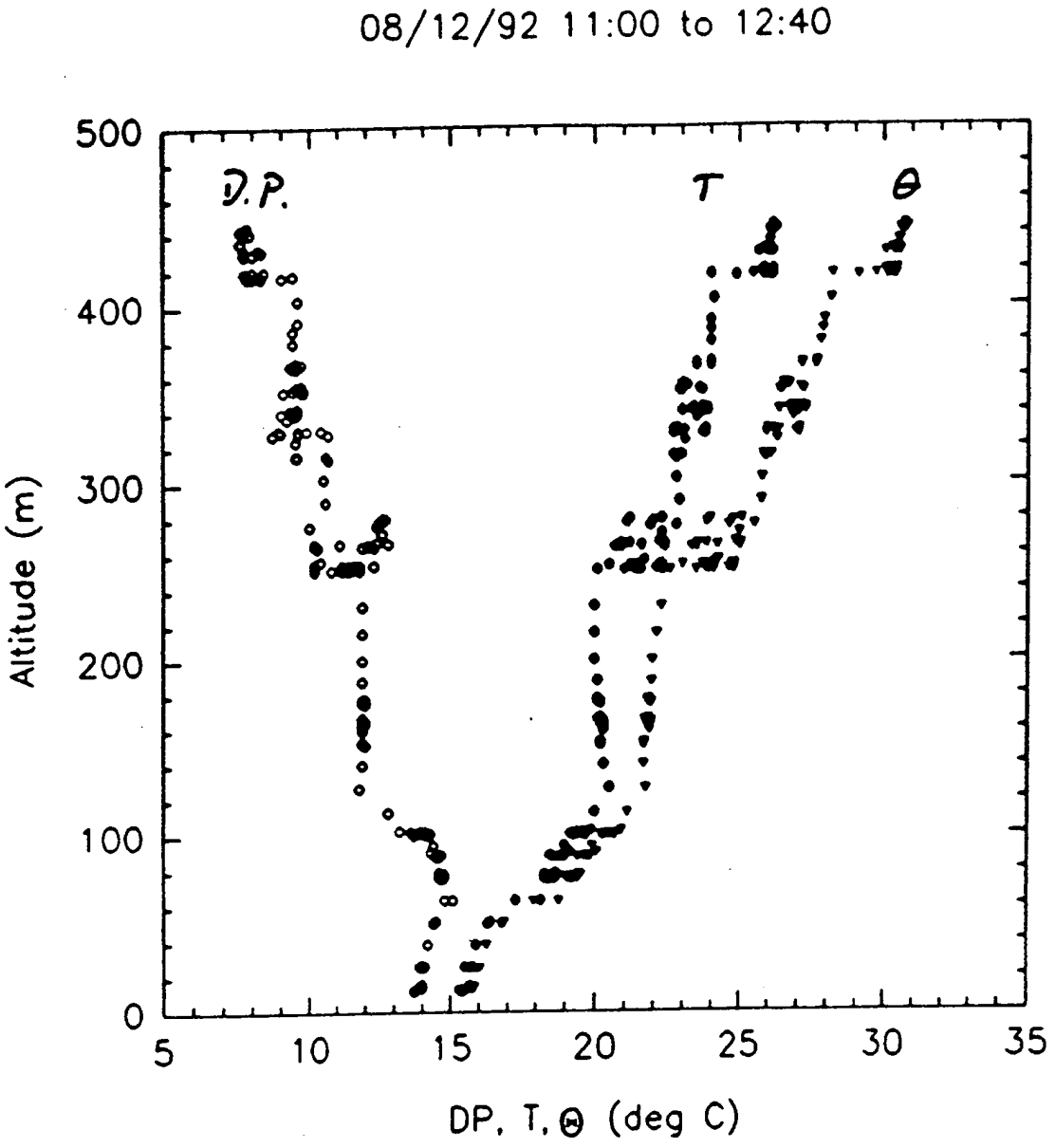


Fig. 3. Nucleation rates of aqueous acid droplets at 25°C at various relative humidities as a function of acid relative acidity. Upper scales: corresponding acid partial pressures in ppm.

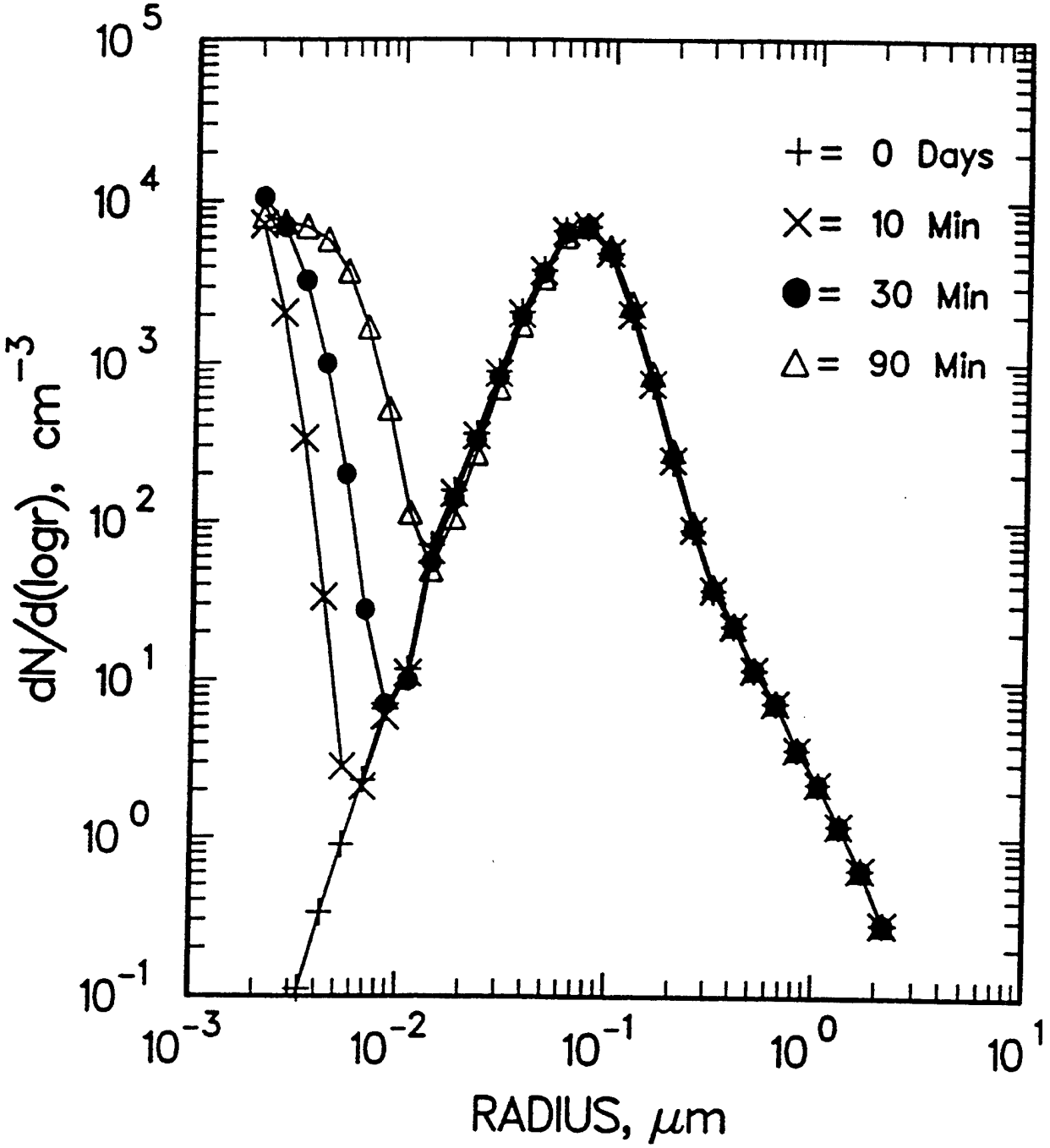
PARTICLE SIZE DISTRIBUTIONS MEASURED ON 12 AUGUST 1992 OFF THE COAST OF OREGON AT AN ALTITUDE OF 185 METERS . DISTRIBUTION AT 13:40 (FILLED CIRCLES) SHOWS WHAT IS BELIEVED TO BE NEWLY NUCLEATED PARTICLES.



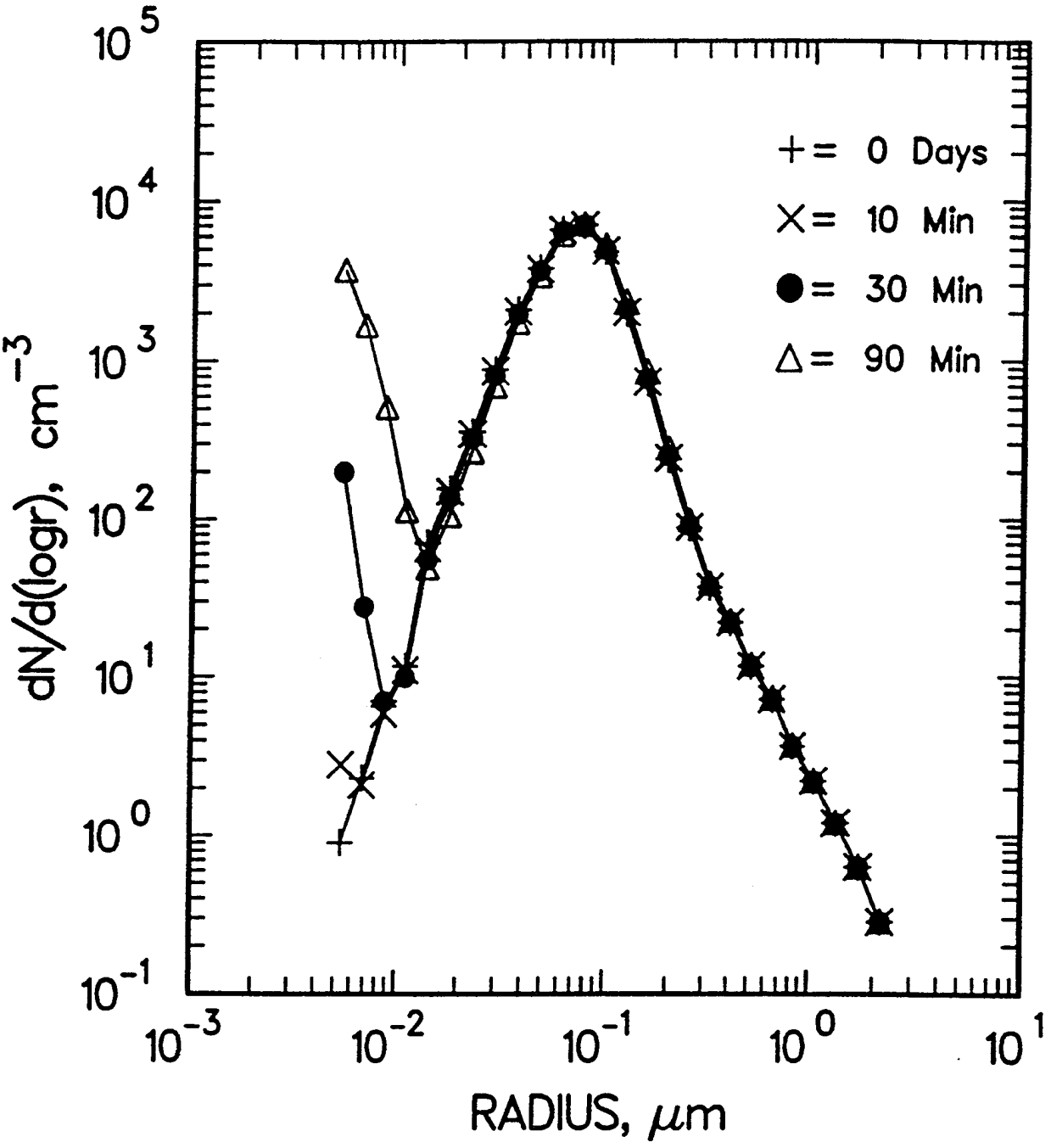
VERTICAL PROFILES OF TEMPERATURE AND DEW POINT AT THE TIME OF THE NUCLEATION EVENT ON 12 AUGUST 1992 OFF THE COAST OF OREGON



MODEL SIMULATION OF THE NUCLEATION EVENT OBSERVED OFF THE COAST OF OREGON ON 12 AUGUST 1992



MODEL SIMULATION OF THE NUCLEATION EVENT OBSERVED OFF THE COAST OF OREGON ON 12 AUGUST 1992



PRECIPITATION SCAVENGING

- Treatment of this process is fairly simple.

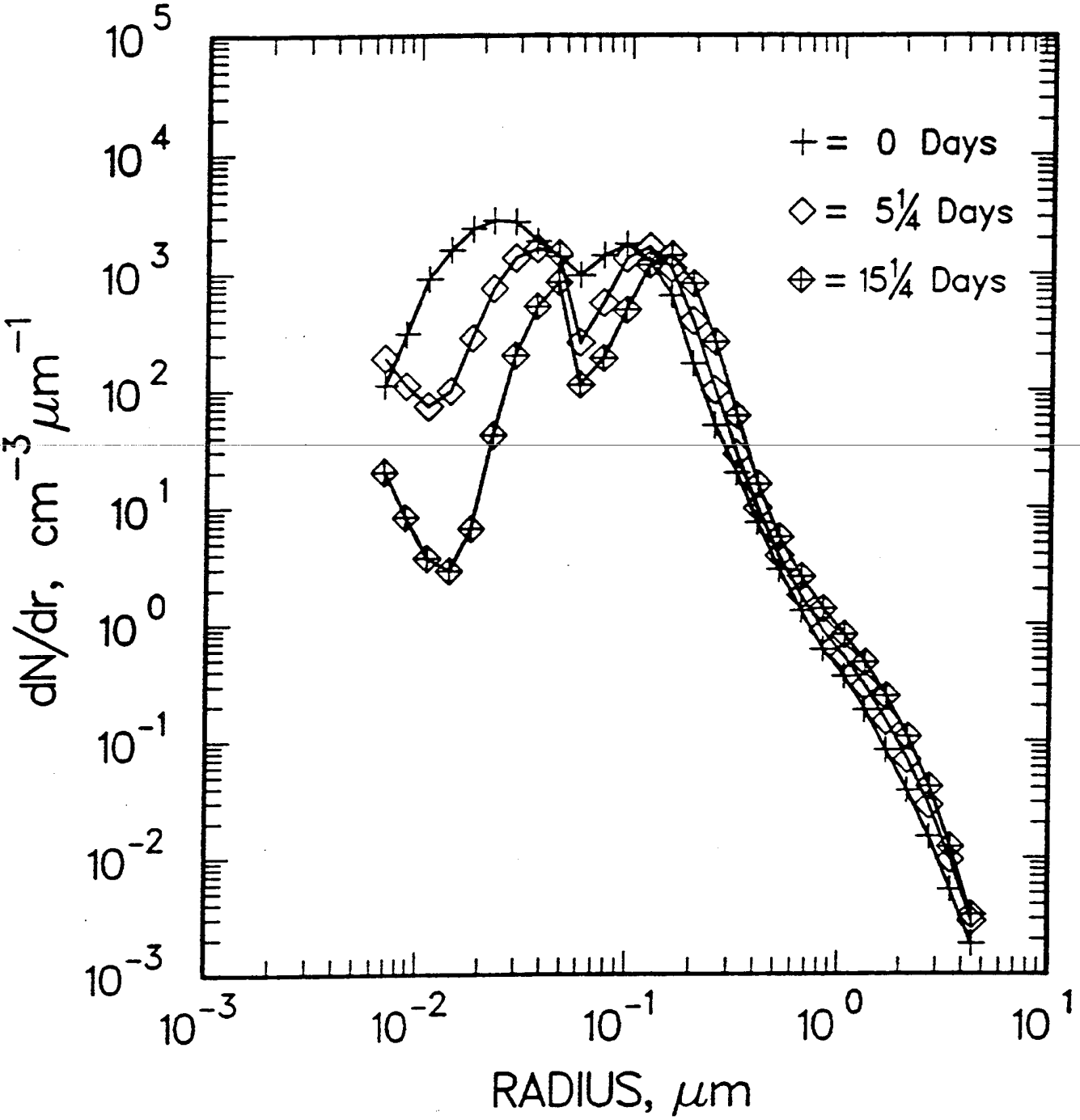
- Presently, we consider only in-cloud scavenging (rainout) of aerosols in those size sections that would be activated in cloud to form cloud droplets. These sections are determined by a specified (inputted) value of the dry size of the smallest particle that will serve as a cloud condensation nucleus (CCN). Rainout occurs in vertical grid levels containing cloud.

- Precipitation scavenging coefficient, Λ (sec^{-1}), used is that estimated by Pruppacher and Klett (1978). This estimate is based on the assumption that rainout is a two-stage process. In the first stage scavenging mechanisms, especially activation (nucleation) of CCN and Brownian diffusion, serve to transfer a significant fraction of aerosol particles to cloud droplets. In the second stage, this "polluted" cloud water eventually ends up on precipitation-sized particles as a result of collision and coalescence between cloud droplets. The accretion process is the rate-limiting step and Pruppacher and Klett take the accretion rate of cloud droplets by precipitation-sized particles as a reasonable upper bound of Λ. This estimate is given by

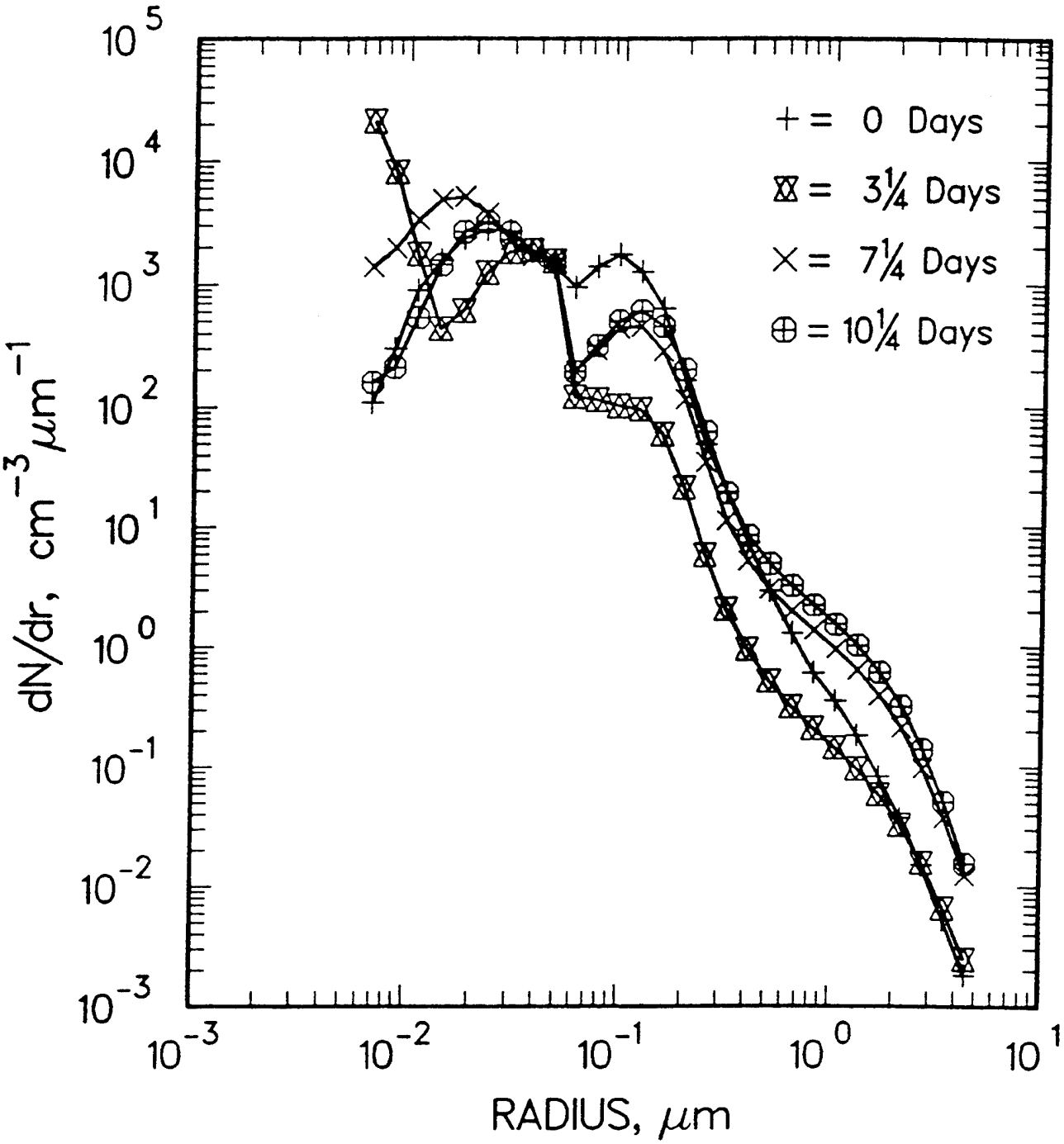
$$\Lambda = 3.49 \times 10^{-4} R^{0.79}$$

where R is rainfall rate in mm/hr. Aerosol mass is removed from all 'activated' sections with the same efficiency, regardless of particle size.

MODEL SIMULATION OF AEROSOL EVOLUTION IN REMOTE MARINE BOUNDARY LAYER SHOWING THAT, IN THE ABSENCE OF PRECIPITATION SCAVENGING, NEW PARTICLE FORMATION BY NUCLEATION WILL NOT BE SUFFICIENT TO MAINTAIN THE OBSERVED CONCENTRATION OF SMALL PARTICLES



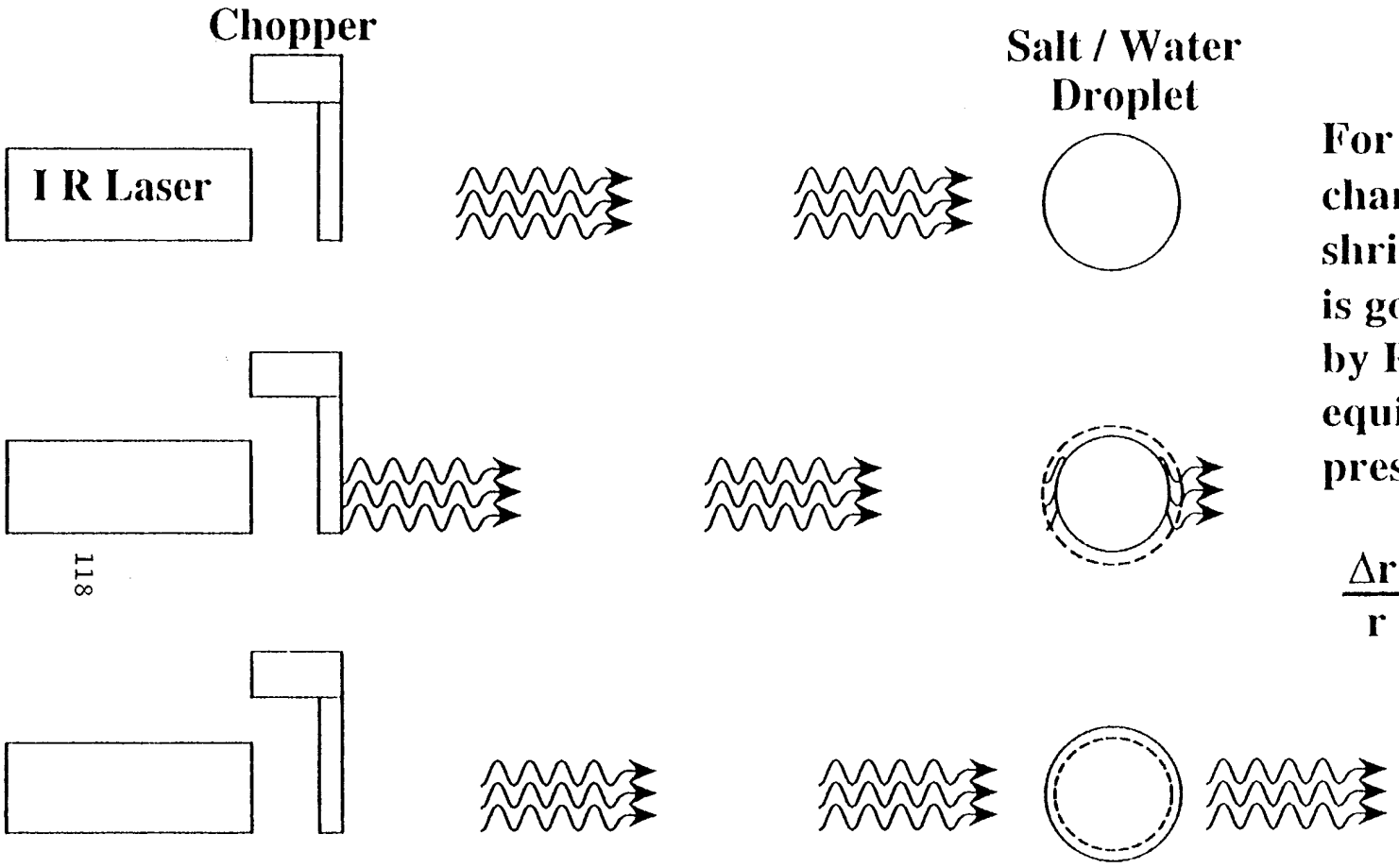
MODEL SIMULATION OF AEROSOL EVOLUTION IN THE REMOTE MARINE BOUNDARY LAYER SHOWING SIGNIFICANT FORMATION OF NEW PARTICLES FOLLOWING PRECIPITATION SCAVENGING OF LARGER PARTICLES



Marine Aerosol Sizing
by
Photothermally Modulated Scattered Light

- Specialized Application to Marine Aerosols
- Novel approach to provide input data and/or verification of aerosol model
- Vertical Profiles of Size Distribution in the first 10 meters possible in moderate to rough seas
- Potential for LIDAR remote sensing technique to obtain aerosol size distributions at range

Photothermal Modulation of Droplet Size

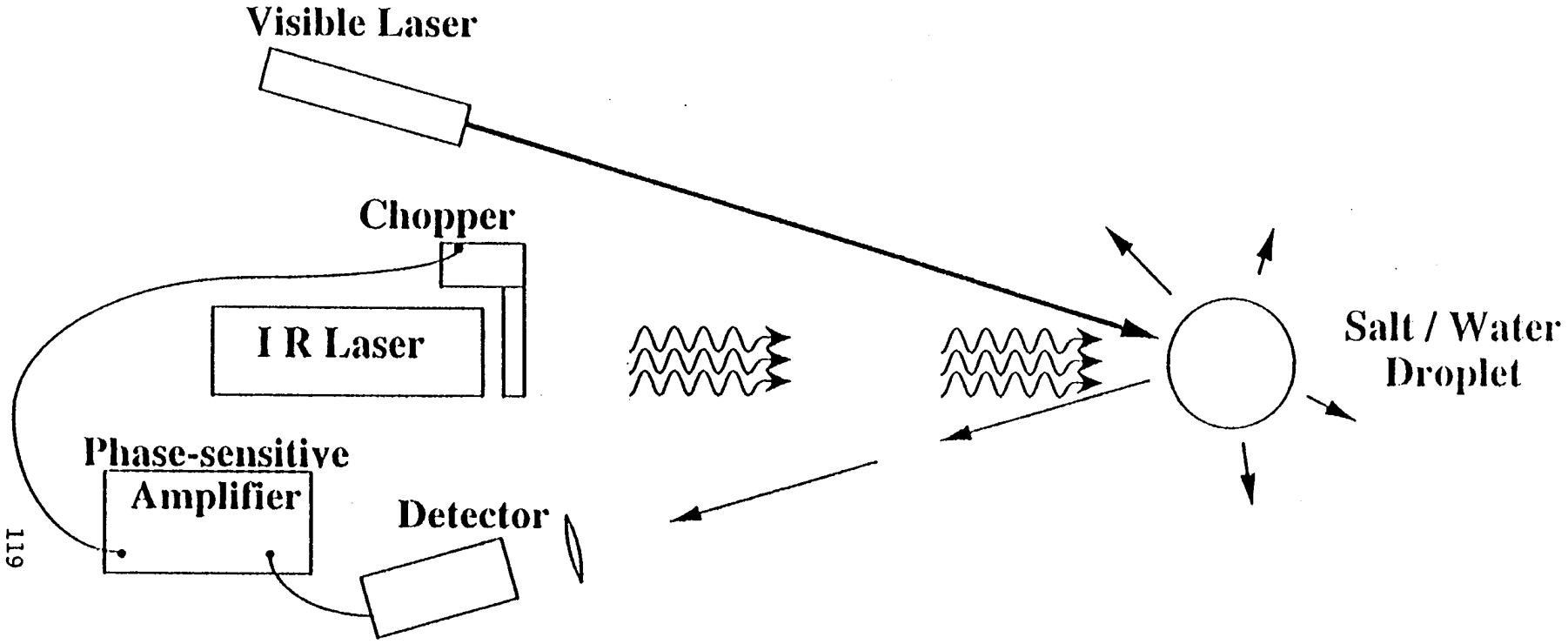


For small temperature changes, droplet shrinkage and regrowth is governed primarily by Raoult's Law for equilibrium vapor pressure :

$$\frac{\Delta r}{r} = -\frac{1}{3}\frac{\Delta T}{T}(1-X)^{-1}$$

Relaxation or response time for size change is a strong function of droplet radius

Droplet sizing using photothermal modulation of scattered light



The nonlinear droplet size dependence on a pulsed IR incident beam can be linearized for small ΔT and Δr. For $r(t) = r_o + \Delta r(t)$, the scattered light signal will be proportional to $\dot{\Delta r}$:

$$\dot{\Delta r} = -\alpha \cdot I - (\gamma_1 / r_o^2 + \gamma_2 \cdot I / r_o) \cdot \Delta r$$

where $\alpha = \alpha(Q, T, X, \rho)$, $\gamma_i = \gamma_i(Q, T, X, \rho)$, and $\gamma_1 > \gamma_2$

For small droplets and/or low IR intensities, γ can be neglected, and the solution is simply [redacted]

$$\Delta r(t) = \frac{\alpha I}{(\omega^2 + \gamma^2 / r_o^4)^{1/2}} \sin(\omega t + \phi)$$

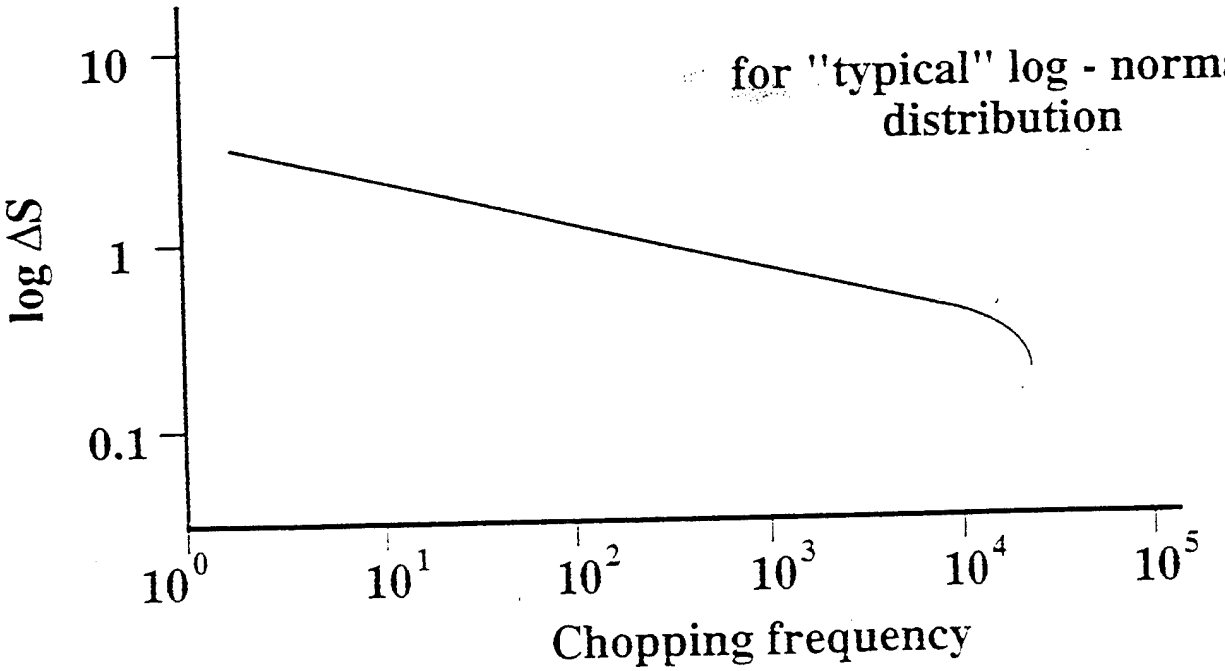
The scattered light signal, ΔS, is porportional to Δr:

$$\frac{\Delta S_\omega(r, t)}{S_\omega} \approx \frac{\Delta r(t)}{r_o} = \frac{\alpha I}{r_o(\omega^2 + \gamma^2 / r_o^4)^{1/2}} \sin(\omega t + \phi)$$

The total signal will be proportional to :

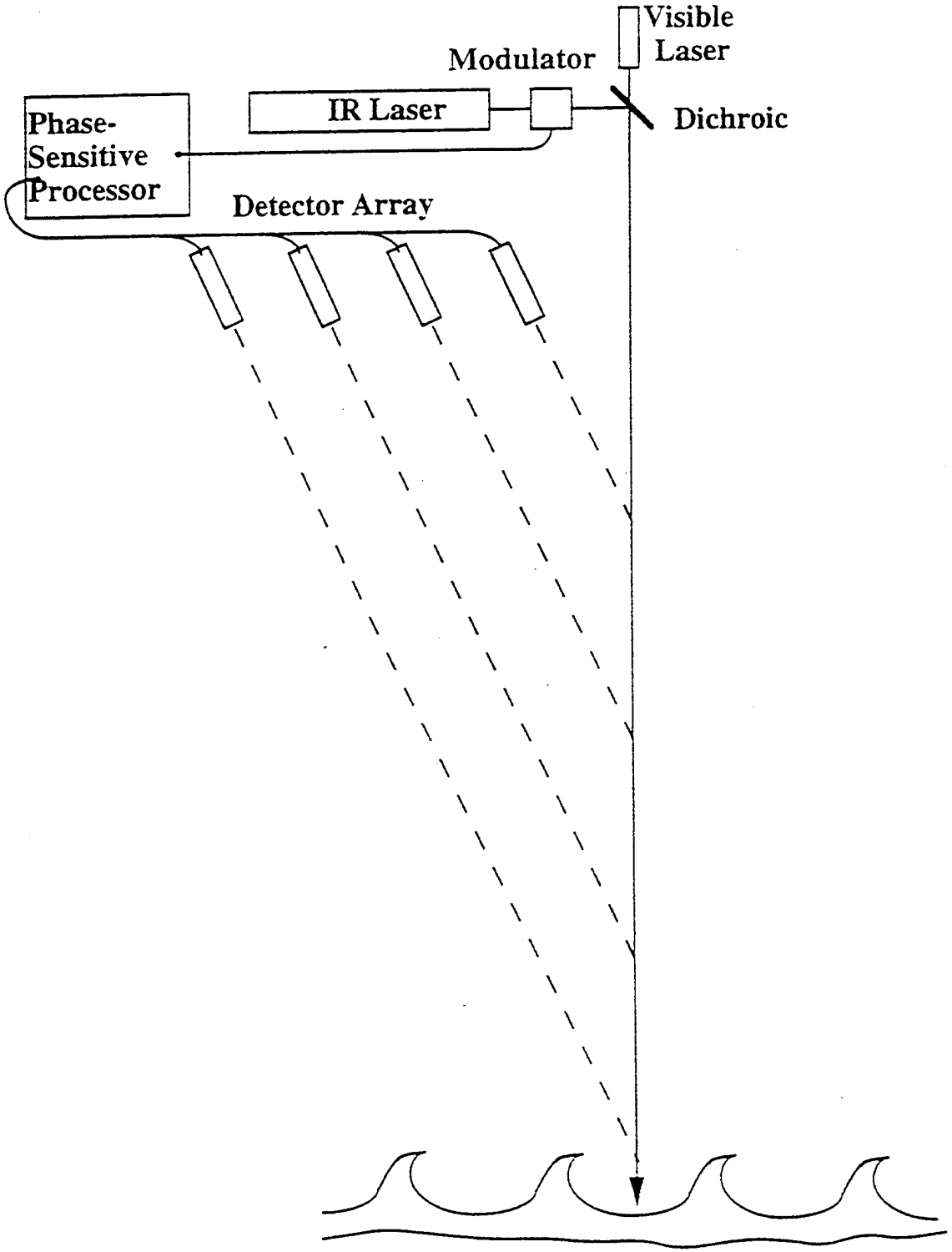
$$\int \frac{n(r_o)\, dr}{r_o(\omega^2 + \gamma^2 / r_o^4)^{1/2}}$$

where n(r) is the size distribution

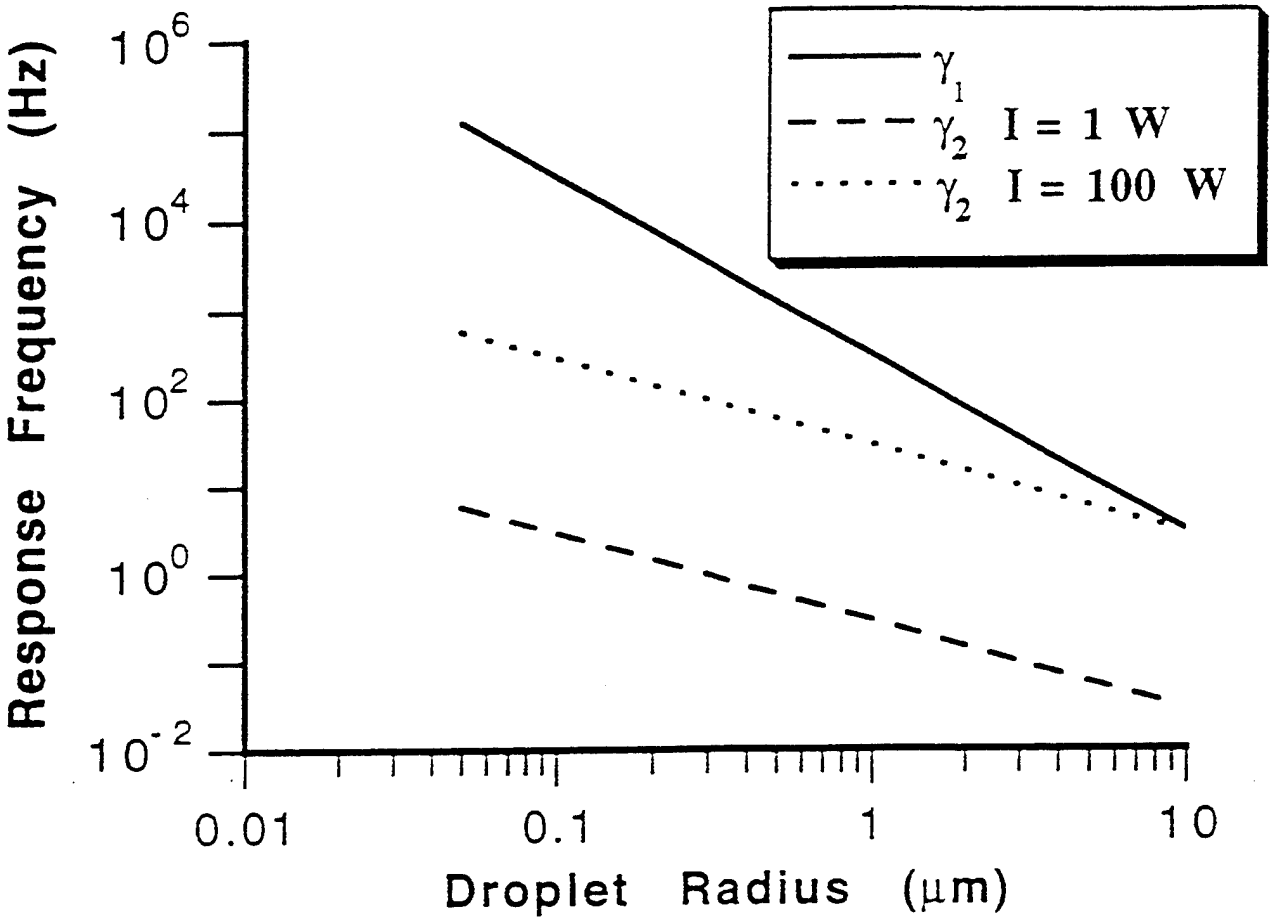


for "typical" log - normal distribution

Vertical Profile Droplet Sizing Using Photothermal Modulation of Scattered Light



HIgher Beam intensities result in non-linear behavior in the return signal for larger droplet sizes



Larger size droplets response can be obtained by signal processing techniques similar to phase fluorimetry

Potential application to droplet size LIDAR

Remote Sensing and Radiative Properties

Deficiencies

•Lack of long term measurement record - Aerosol Properties (Size Distribution, Optical Depth, Single Scatter Albedo, Sources)

•Characterization of Source Regions

•Spatial Resolution Required - Vertical - 1 to 10 m; Horizontal - 100 to 1000m

•Aerosol Models(for variability within coastal environment) - Sea Salt, H_2SO_4, dust (composition, size, shape), n(r), w_0, R.I. (Rel Hum), Organics, Mixed aerosol (heterogeneous vs. homogeneous)

•Characterization of water leaving radiance - sun glint, sea state, wind speed, subsurface (biogenic, sediment)

•Mie scattering for non-spherical aerosol, heterogeneous particles

•Inclusions of multiple scattering effects

•Errors in applying inversion techniques

•Closure measurements / Complete radiation experiment

Remote Sensing and Radiative Properties

Solutions

•Combination of model, in situ, and remote sensing observations

•Closure experiments - (Judge models and measurements), Combine with other programs to leverage Navy interests (TARFOX, ACE 1, ACE 2

•Lidar shipboard and ground based

•Measurements for process studies/ radiative properties

•Measurement for real time analysis/Inputs for modeling

•Other operational measurements - Nephelometers, Sun photometers, Transmissometers (As function of λ)

•UAV's

Remote Sensing and Radiative Properties

Consequences Without Remote Sensing

- Lack of large scale coverage
- Miss uncharacterized sources
- Limited information in data denied areas (hostile environments)

Remote Sensing and Radiative Properties

Other Issues and Needed Coastal Meteorology Effects

- Relative Humidity - need high accuracy near saturation
- Precipitation - Remote estimates and Modeling

Short term mesoscale forecasting - Aerosol, clouds, fog

Remote Sensing and Radiative Properties

Recommendations

- Extend monochromatic and bispectral aerosol retrievals to multi-spectral and hyperspectral
- Include multiple scattering effects
- Treatment of water leaving radiance
- Lidar for real time analysis/ input support of process studies
- Closure experiments
- Remote sensing of inputs for models - sea state, whitecaps, available actinic radiation, troposphere O_3, SO_2

Sea Surface Aerosol Source Function

Current State of the Art

1.Bubble-generated aerosols: Source functions known to ~1 order of magnitude - Look for closure within 5-10 years

2.Spume droplet production: Source function not known to within 3 order of magnitude: Wind dependence, U_*^3; Understanding ~1 decade behind 1.

3. Initial estimates suggest that surf zone will be significant (>10%) source of coastal zone aerosols.

4. We need experiments (lab and field) of bubble populations as well as aerosol populations to inform our understanding of the physics of bubble mediated aerosol production, e.g., the efficiency of droplet production (drops/ bubbles) in open-ocean conditions, surf zone, and the mechanics of jet-droplet production, ...

5.Near surface aerosol distributions significantly modified by wave field

6.Surface films and high concentrations of bulk organics - both prevalent in coastal waters, will profoundly modify sea-surface aerosol production.

7.Rain induced droplets (splash and via intermediate bubbles) may be episodically very important - Are the splash droplets strictly fresh water?

Spatial variability (horizontal) Greater in coastal regions than in open ocean

Sea Surface Aerosol Source Function

Aerosol Sink Function

The frequency of wave breaking (whitecapping) and the extent of surface organic films will alter the effective "dry" deposition velocities of aerosols (anthropogenic as well as marine background)

Sea Surface Aerosol Source Function

Recommendations

1. Need to study (in field and laboratory) specific mechanisms that generate spray droplets in surf and other coastal zones.

2. Need to study, in field, production of spume droplets by natural(wave breaking) mechanisms.

3. Need detailed investigations of bubble flux/droplet flux in breaking wave (field and lab) conditions

4. Develop finite list of parameters (remotely monitorable, including beach noise, meteorology, and satellite ..) required for aerosol nowcast.

Physics and Chemistry of Coastal Aerosols

State of the Art

- Source issues

  - Nucleation of new particles - SO_2, DMS, MSA, (NH_3), (H_2O) in marine atmosphere

  - MBL entrainment issues - Aerosol as a tracer, Mass($\tau \sim$ hours)

  - Precursor gases

Physics and Chemistry of Coastal Aerosols

Direct Injection

What are the size resolved aerosol chemistry and physics and optical properties for the following sources

- Biomass burning (high temp)
- Desert dust
- Urban plume
- Biological organics (low temperature)
- Sea surface
- Emissions of gases

Physics and Chemistry of Coastal Aerosols

Nucleation

- What, besides sulfur, are the chemical species nucleating new particle

- What is the dominant mechanism (binary, ternary, ion- induced ...) for new particle formation?

- What environmental conditions (thermodynamic/chemical) necessary for new particle formation (e.g. BL vs. free troposphere)

Physics and Chemistry of Coastal Aerosols

Cloud Processing

• To what extent does particle composition and size affect droplet nucleation and interstitial aerosol?

•What is the relative importance of the various oxidants in the production of sulfuric acid?

•Can oxidation in cloud droplets be described by the "bulk" chemical composition of individual droplets

•What is the role of entrainment and turbulence in the processing of aerosols in clouds?

Physics and Chemistry of Coastal Aerosols

Heterogeneous Processes

- What are the condensation coefficients for the major condensing species?
- What are the important chemical reactions on aerosol particles?

Physics and Chemistry of Coastal Aerosols

Sinks

•What is the precipitin scavenging efficiency of aerosol particles as a function of size and composition?

• How adequate are the present dry deposition algorithms of aerosols and gases to the sea surface?

Physics and Chemistry of Coastal Aerosols

Laboratory Studies

- Homogeneous chemistry - Determine dominant oxidation pathways and rate constants for DMS and SO_2
- Test proposed nucleation mechanisms
- Cloud processing - Test theories
  - SO_2 by O_3 and H_2O, ...
  - Neutralization by NH_3
  - Rates
- Investigate relative importance of proposed heterogeneous reaction schemes
- **Need integrated aerosol/air chemistry/cloud physics expertise and participation**
- **In each case, test model of process against lab result**

Physics and Chemistry of Coastal Aerosols

Field Measurements

•Need size resolved aerosol physics and chemistry for the following source regions:

- •Biomass burning
- •Desert dust
- •Urban plume
- •Biological source
- •Sea surface.

•Need field measurements to define environmental conditions under which new particle nucleation occurs. Consider new particle formation vs. entrainment.

•Lagrangian experiment in air masses advecting off the East Coast.