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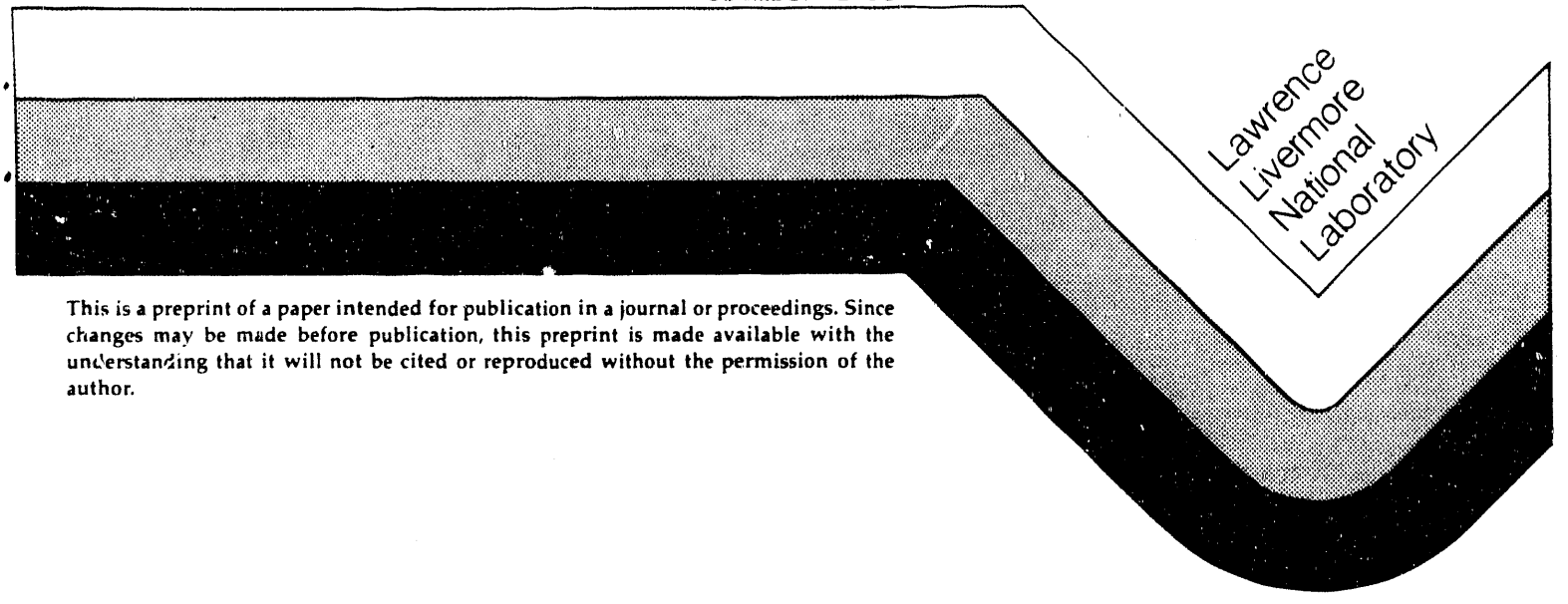

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The Relationship Between Aerosol and Drop Size Distributions in the Marine Atmosphere

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I. Introduction

The characteristics of droplet size distributions near cloud base are initially determined by the aerosol particles that act as cloud condensation nuclei (CCN). Through physical and chemical processes that determine the fraction of aerosol particles that act as CCN, these particles profoundly affect the microphysical structure of clouds and their tendency to form precipitation.

This paper provides a parametric study of the condensational growth on aerosol distributions from typical measurements in the marine atmosphere, and answers the following questions with respect to this process.

(1) How does the condensationally produced droplet spectrum vary with the initial aerosol size distribution, aerosol number loading, and the updraft velocity?

(2) What is the fraction of aerosol particles that act as CCN? This information can be applied to the future study to parameterize the number concentration of cloud droplets in climate models.

(3) How do the optical properties of the cloud change as a result of the production of different droplet size distributions?

2. Microphysical Model

We use a dynamic framework that describes a well-mixed, spherical air parcel with coupled warm rain microphysics. This model computes the spectral evolution of interstitial aerosols and cloud droplets with time by solving the atmospheric microphysical equations (Pruppacher and Klett, 1978). The model assumes adiabaticity because we are interested only in the initial stages of cloud development. We examine the consequences of diffusional growth on an aerosol (ammonium sulfate, $(NH_4)_2SO_4$) distribution which consists of a nucleation and an accumulation mode as shown in Fig. 1.

3. Results

a. Effect of aerosol number loading

Fig. 2 shows three aerosol distributions which correspond to an increase of total number density from 100 to 1000 cm^{-3}. Fig. 3 presents the resulting distributions of unactivated aerosol and cloud droplet for these three aerosol number loadings and different updraft velocities at 50 m above the cloud base. The growth of cloud droplets by water vapor diffusion is more rapid for lower initial aerosol concentration, thereby leading to a narrow spectrum and a clear separation between aerosols and droplets. In addition, the lower aerosol loading results in a droplet size distribution that is more likely to be bimodal at higher velocity, although the amount of available water vapor limits the growth of droplets to a somewhat smaller size.

b. Effect of initial aerosol distribution

The measured aerosol distribution in Fig. 1 can be described with the sum of two normal distributions. We changed the relative magnitude at each mode but maintained the total number a constant. The resulting three aerosol distributions are given in Fig. 4. The corresponding droplet spectra under the same conditions as Fig. 3 but for aerosol distributions b and c of Fig. 4 are illustrated by Fig. 5a and 5b. We found that the different initial aerosol distributions given in Fig. 4 do not create significantly different droplet size distributions except for the case with the lower aerosol number concentration and the higher updraft velocity.

c. Fraction of aerosol particles activated to droplets

Fig. 6 presents the fraction of the total aerosol number that is activated to cloud droplets as a function of updraft velocity for the three types of initial aerosol distributions given in Fig. 4 and variable aerosol number loadings. As shown in Fig. 6, for low initial aerosol number concentrations the different initial aerosol distributions only affect the nucleation fraction at lower updraft velocities. However, this fraction strongly depends on both the quantity of aerosol concentration present at cloud base and the updraft velocity. As the velocity is increased, the rate of condensation cannot respond quickly enough to compensate for the increased cooling rate of the rising air parcel, creating a higher saturation ratio and higher droplet concentration. This figure also shows a higher fraction for smaller initial aerosol loadings. The fewer number of aerosol particles available for nucleation causes a decrease in condensation and an increase in the saturation ratio, thereby leading to a higher nucleation fraction.

d. Variation of the optical properties of clouds

The optical depth of the cloud is proportional to the quantity of $\Sigma a_i^2 \Delta N_i$ (= A), where a_i is the radius of the droplets and ΔN_i is the number of droplets of size a_i. Fig. 7 shows the optical properties of the predicted droplet spectrum for all the cases considered in Fig. 6. Since the droplet number N is a key factor in determining the optical depth, the value of A increases with the increase of aerosol number loading and the updraft velocity. By varying the drop size distribution, the albedo of clouds may be changed, thereby altering the scattering and absorption of solar radiation.

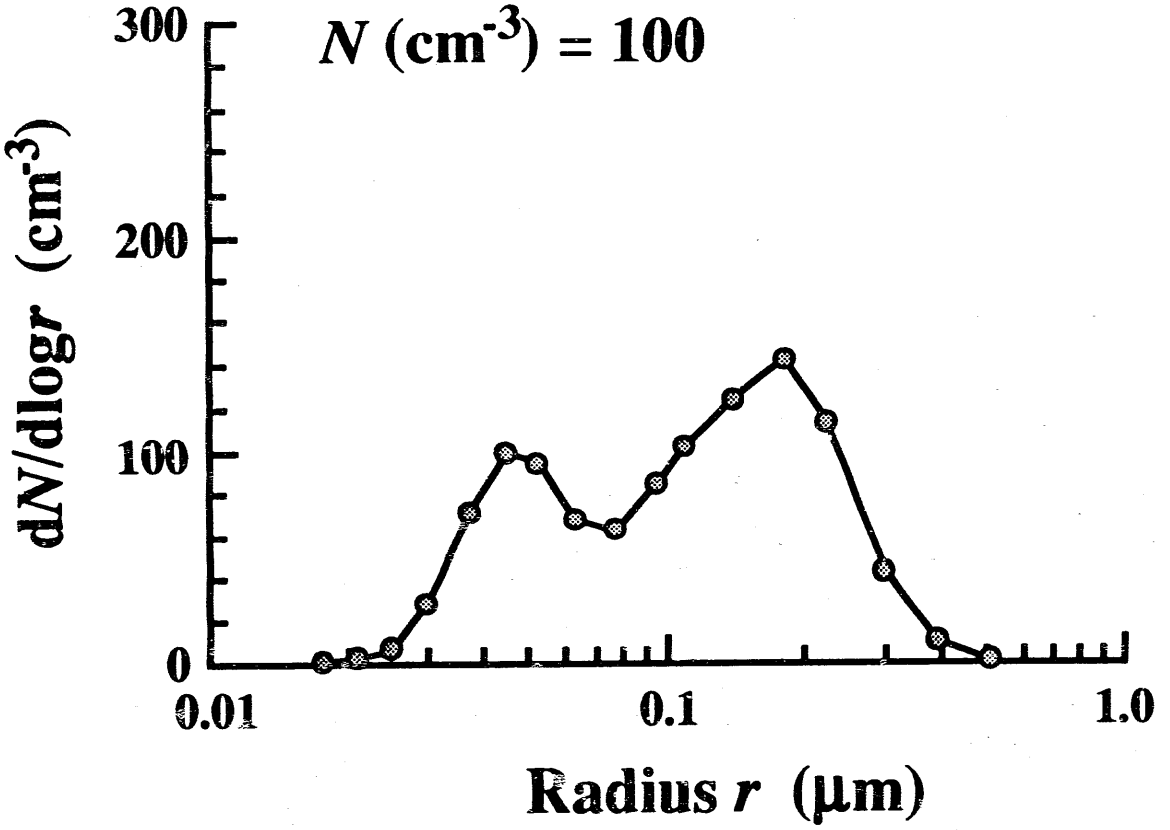

Figure 1

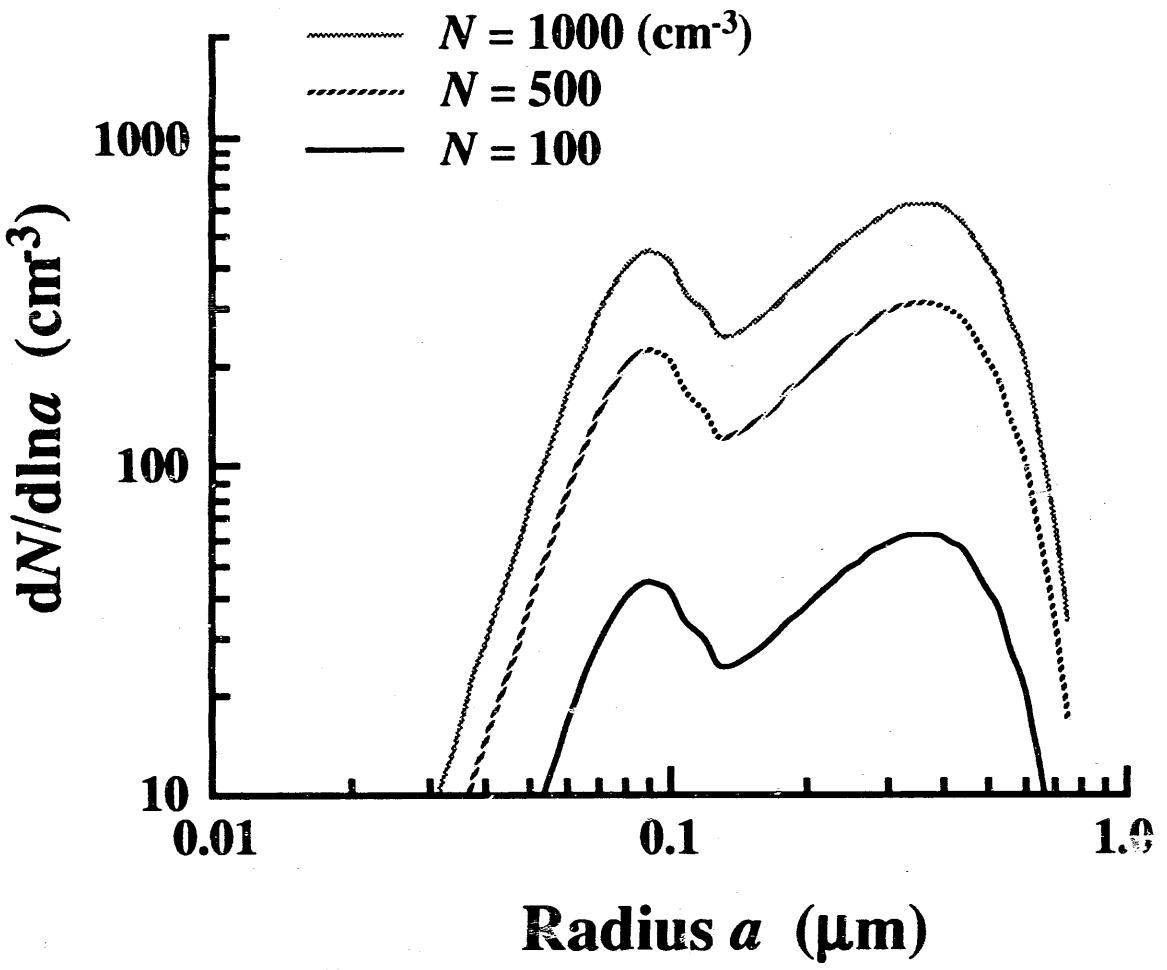
N = 1000 (cm-3)
N = 500
N = 100
1000
100
10
dN/dlna (cm-3)
0.01
0.1
1.0
Radius a (μm)

Figure 2

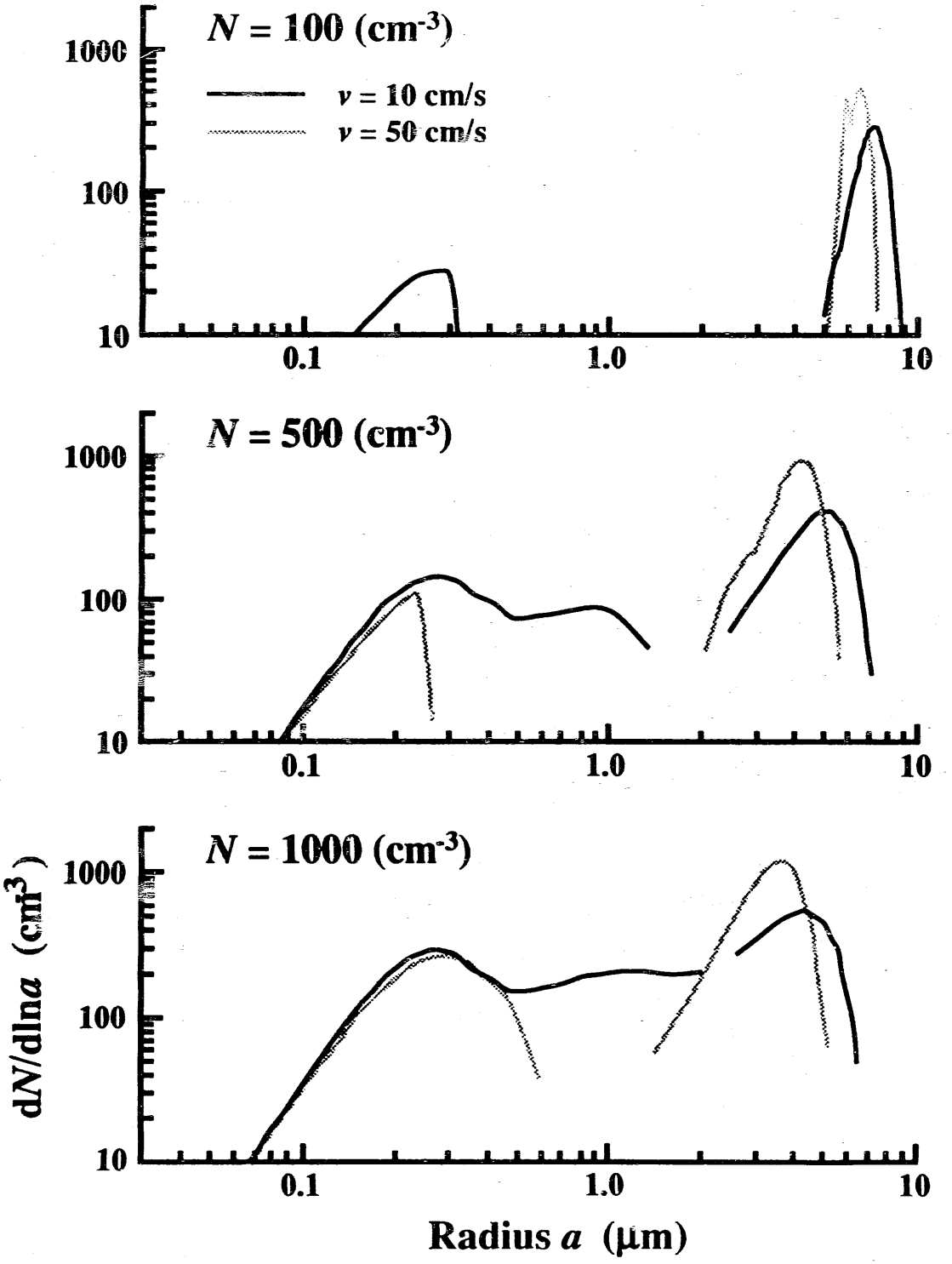
N = 100 (cm-3)
v = 10 cm/s
v = 50 cm/s
1000
100
10
0.1
1.0
10
N = 500 (cm-3)
1000
100
10
0.1
1.0
10
N = 1000 (cm-3)
dN/dlna (cm-3)
1000
100
10
0.1
1.0
10
Radius a (μm)

Figure 3

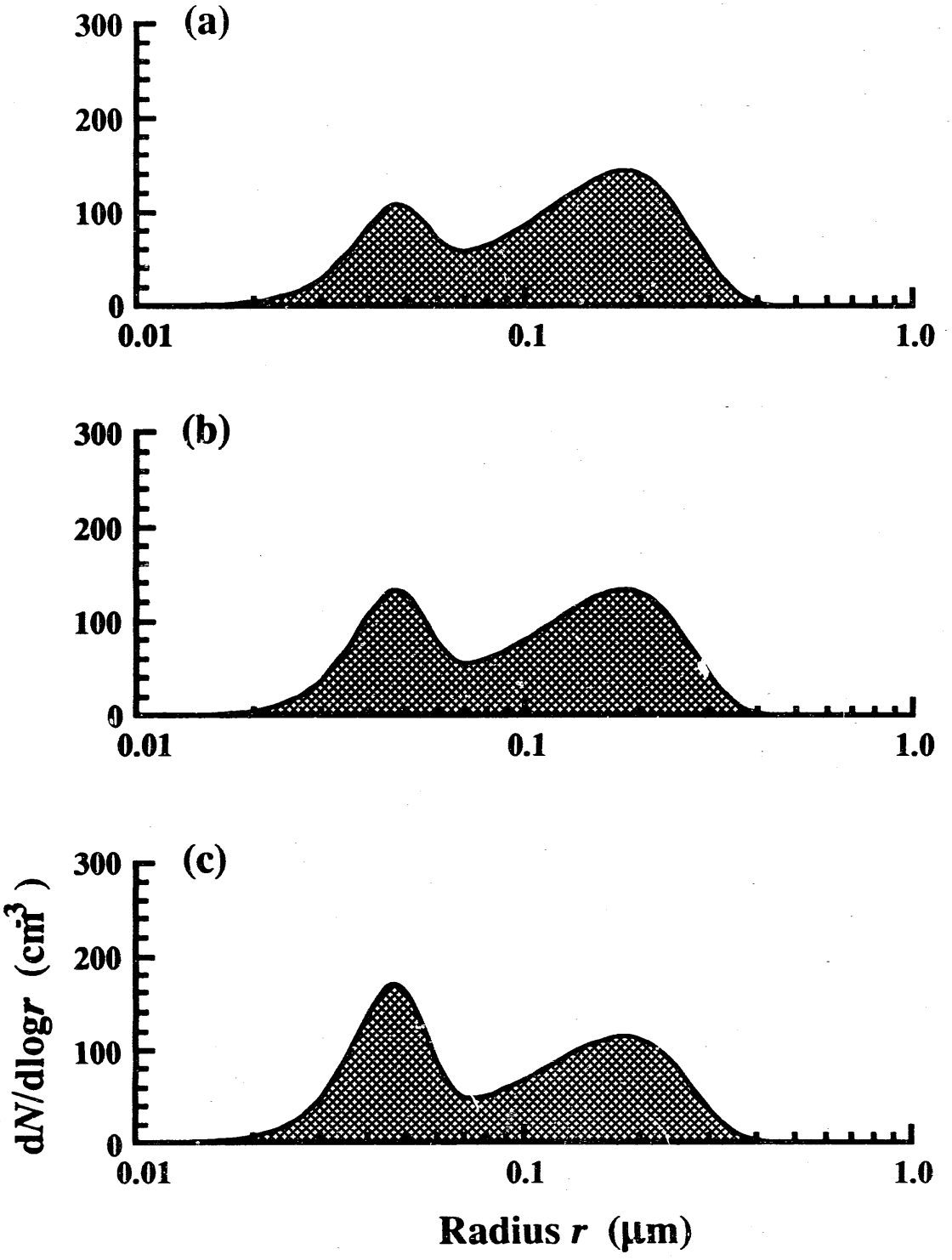
(a)
300
200
100
0
0.01
0.1
1.0
(b)
300
200
100
0
0.01
0.1
1.0
(c)
300
200
100
0
0.01
0.1
1.0
dN/dlogr (cm-3)
Radius r (μm)

Figure 4

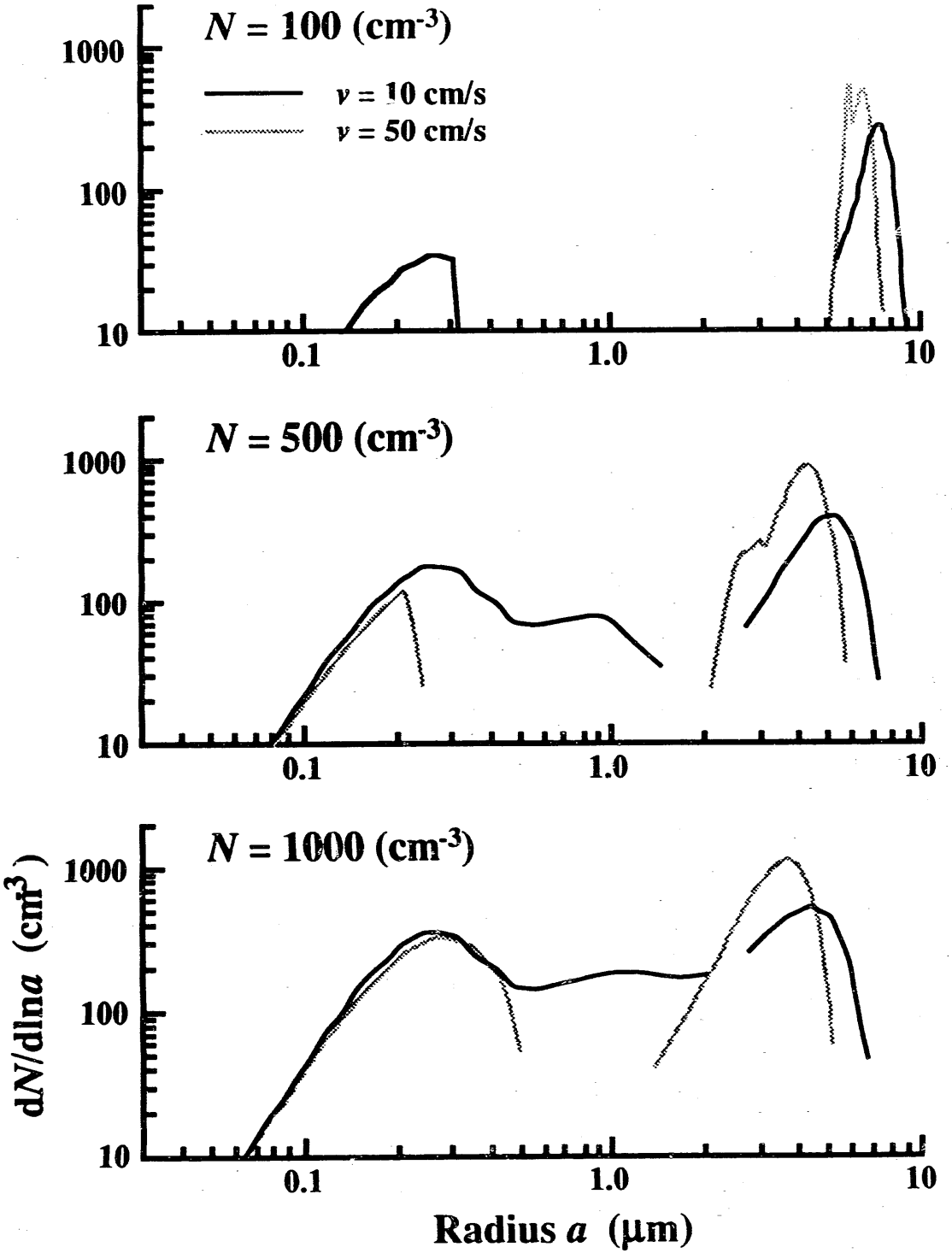
N = 100 (cm-3)
v = 10 cm/s
v = 50 cm/s
1000
100
10
0.1
1.0
10
N = 500 (cm-3)
1000
100
10
0.1
1.0
10
N = 1000 (cm-3)
1000
100
10
0.1
1.0
10
dN/dlna (cm3)
Radius a (μm)

Figure 5a

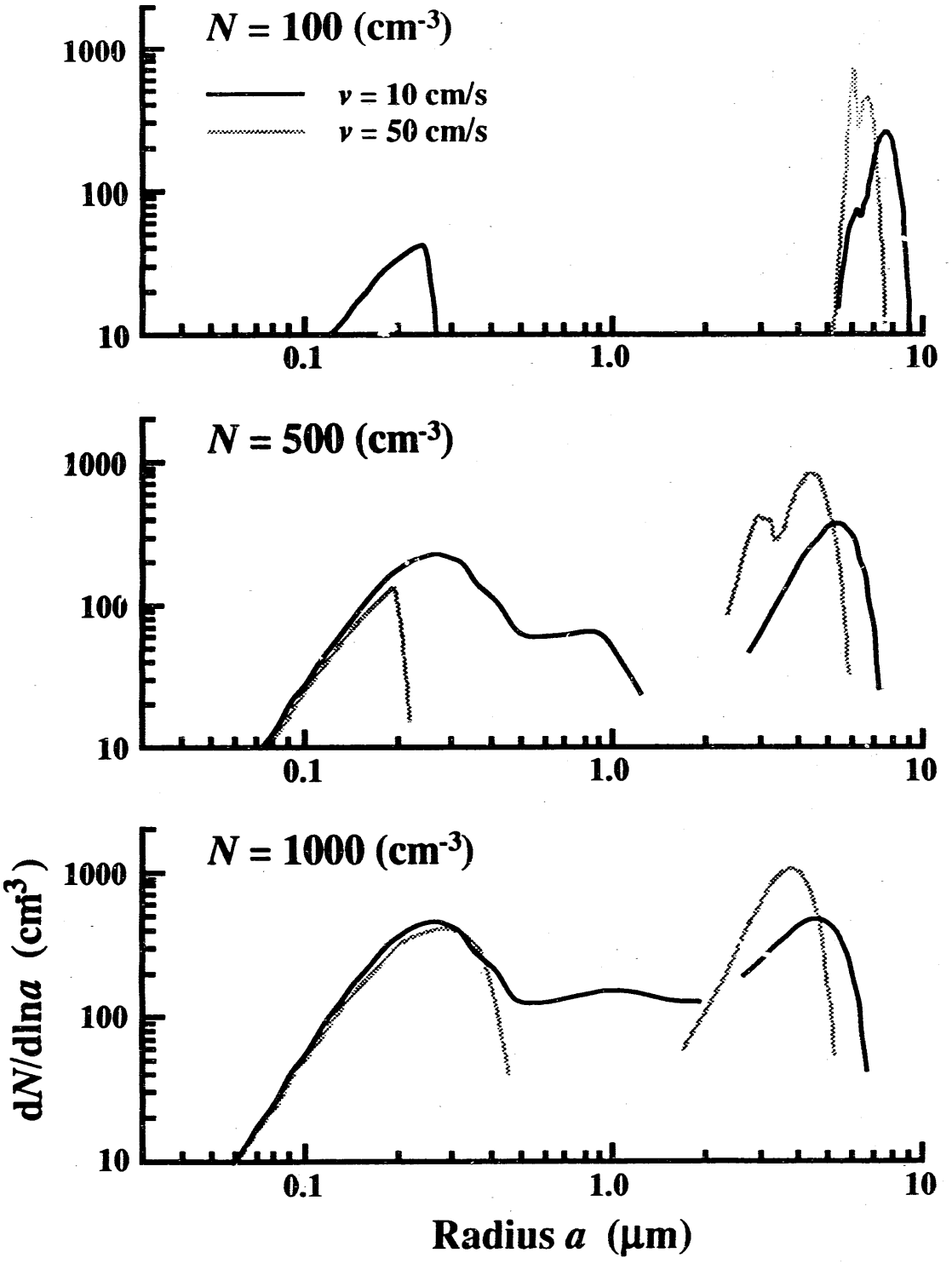
N = 100 (cm-3)
v = 10 cm/s
v = 50 cm/s
1000
100
10
0.1
1.0
10
N = 500 (cm-3)
1000
100
10
0.1
1.0
10
N = 1000 (cm-3)
1000
100
10
0.1
1.0
10
dN/dlna (cm3)
Radius a (μm)

Figure 5b

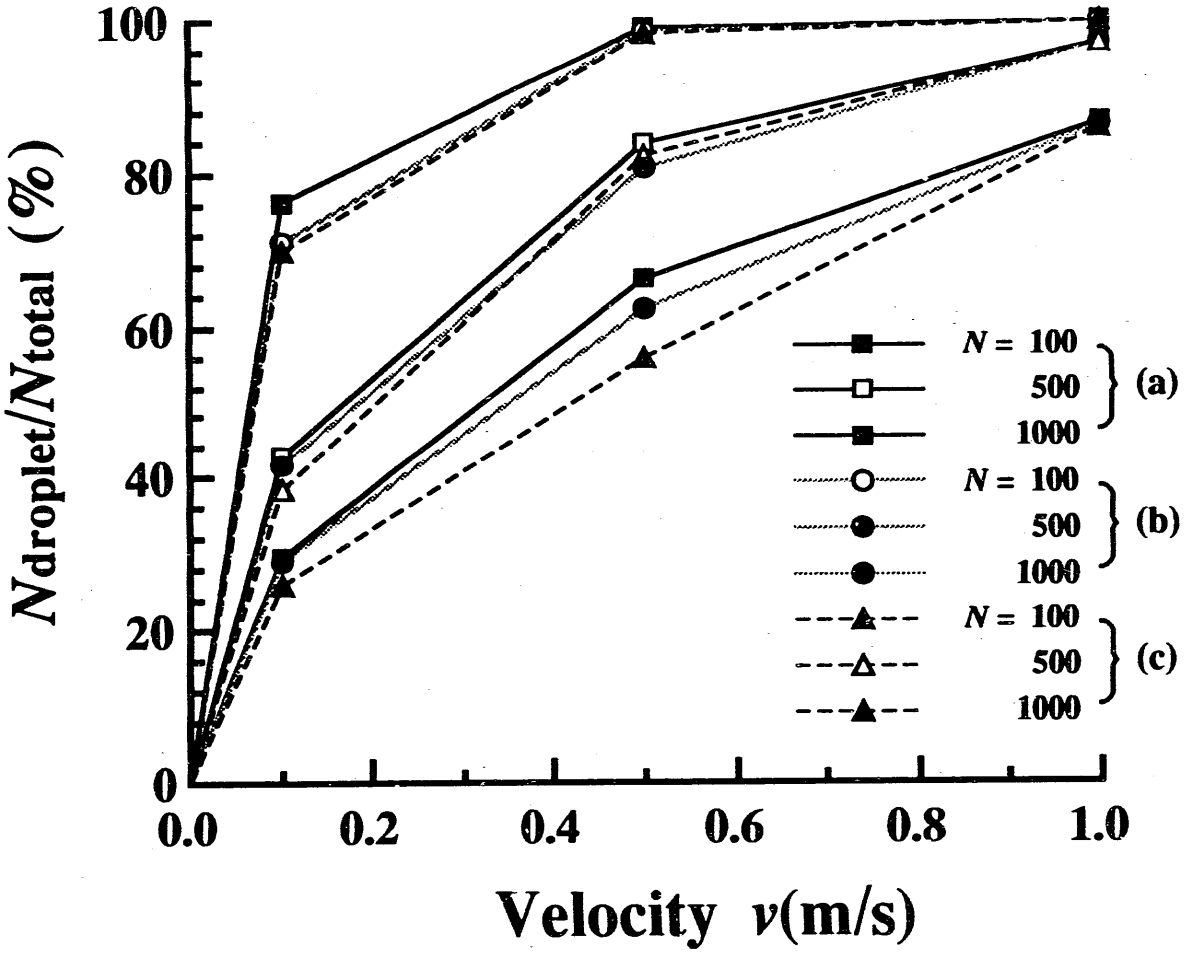
100
80
60
40
20
0
0.0
0.2
0.4
0.6
0.8
1.0
Ndroplet/Ntotal (%)
Velocity v(m/s)
N = 100
500
1000
(a)
N = 100
500
1000
(b)
N = 100
500
1000
(c)

Figure 6

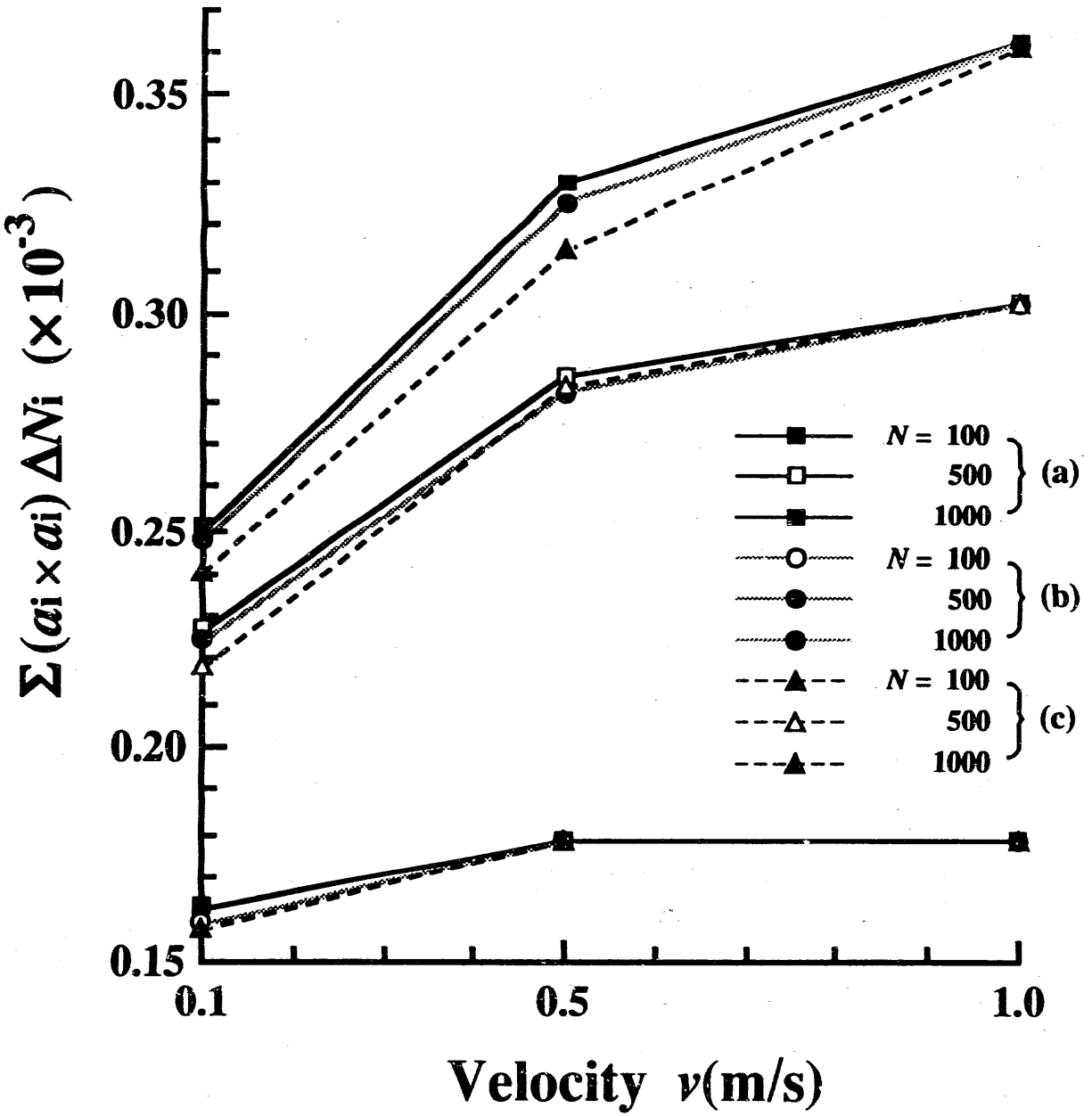
0.35
0.30
0.25
0.20
0.15
0.1
0.5
1.0
Σ(αi × αi) ΔNi (×10-3)
Velocity v(m/s)
N = 100
500
1000
(a)
N = 100
500
1000
(b)
N = 100
500
1000
(c)

Figure 7

4. Conclusions

A detailed microphysical Lagrangian parcel model has been used to study the relationship between measured maritime aerosol particles and the condensationally produced drop size distributions. In this model, the drop number concentration is related to the chemical composition of the aerosol particles, their size distribution, total aerosol mass loading, and updraft velocity.

Results from this study will be used to evaluate the accuracy of a prescribed monodisperse drop size distribution in climate models, and to provide a background for the parameterization of drop size distributions in the study of the Earth's radiation budget.

5. Acknowledgments

Work performed under the auspices of U.S. Department of Energy by the Lawrence Livermore National Laboratory under contract No. W-7405-ENG-48.

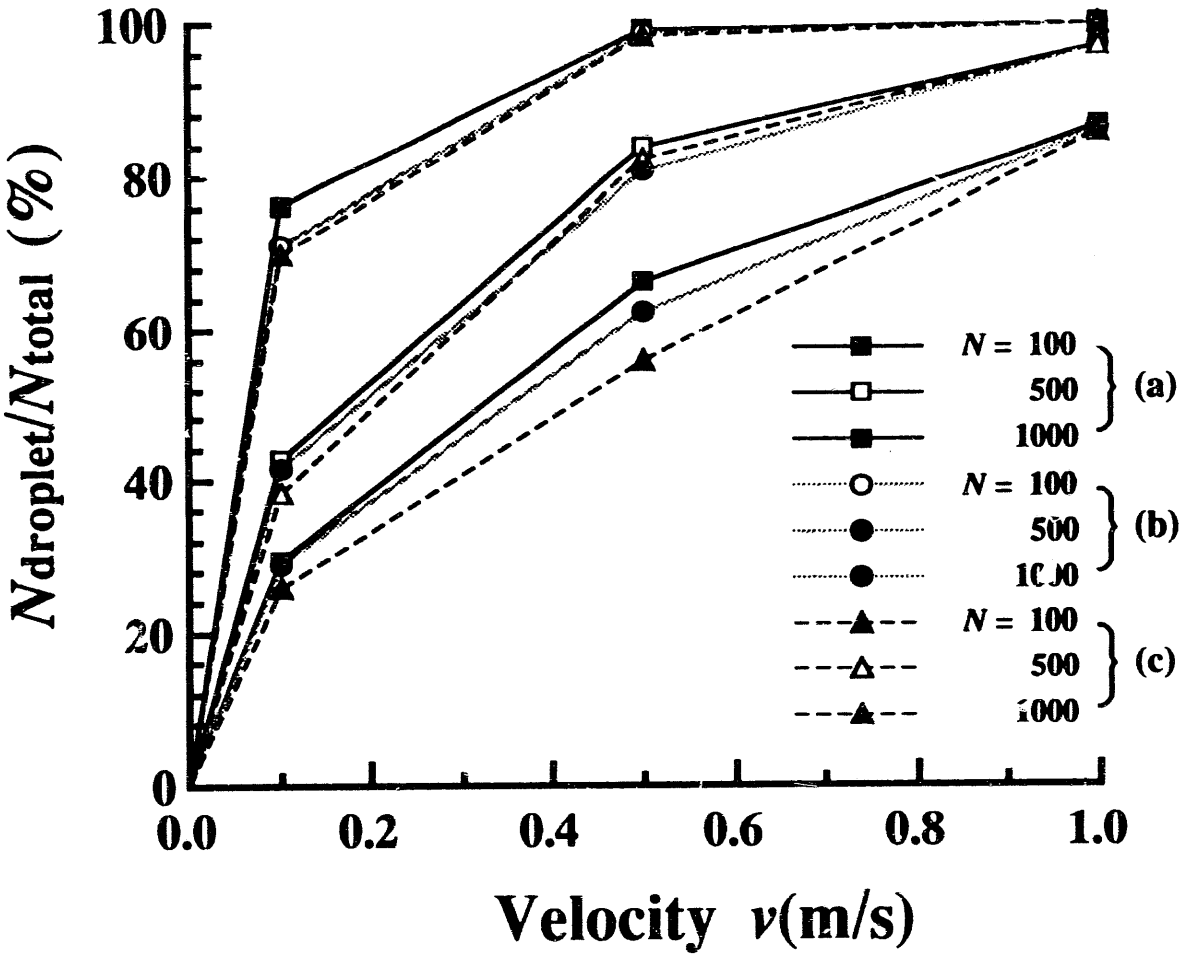
100
80
60
40
20
0
0.0
0.2
0.4
0.6
0.8
1.0
Ndroplet/Ntotal (%)
Velocity v(m/s)
N = 100
500
1000
(a)
N = 100
500
1000
(b)
N = 100
500
1000
(c)

Figure 6

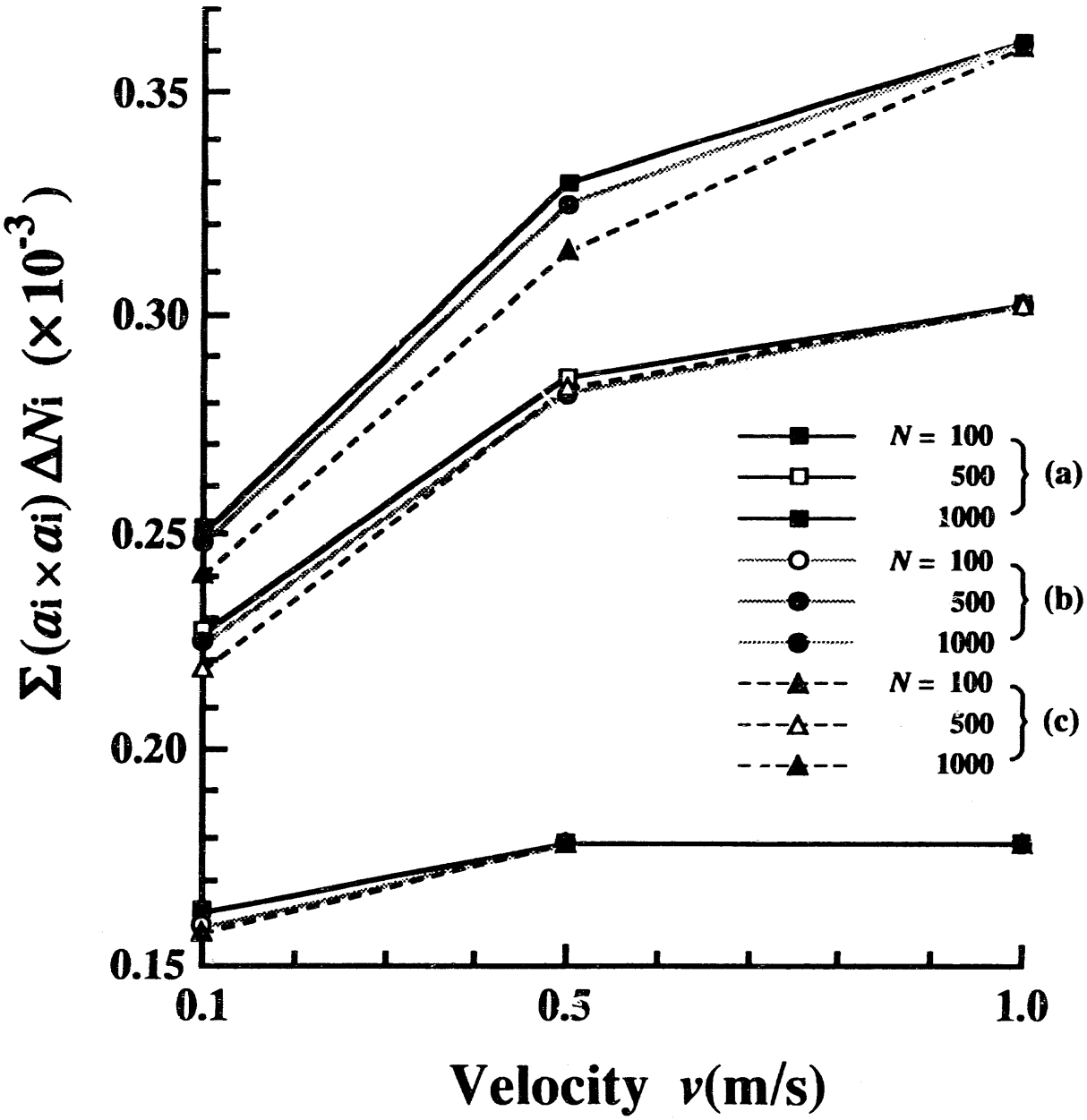
0.35
0.30
0.25
0.20
0.15
0.1
0.5
1.0
Σ(ai × ai) ΔNi (×10-3)
Velocity v(m/s)
N = 100
500
1000
(a)
N = 100
500
1000
(b)
N = 100
500
1000
(c)

Figure 7

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