BNL--47743

DE93 001981

FRACTIONAL ACTIVATION OF ACCUMULATION-MODE PARTICLES IN WARM CONTINENTAL STRATIFORM CLOUDS

N. V. Gillani*, S. E. Schwartz, and P. H. Daum
Environmental Chemistry Division
Department of Applied Science
Brookhaven National Laboratory
Upton, NY 11973

and

W. R. Leaitch, J. W. Strapp, and G. A. Isaac
Atmospheric Environment Service
Downsview, Ontario, Canada

July 1991

For inclusion in
Proceedings of the Fifth International Conference on
Precipitation Scavenging and Atmospheric-Surface
Exchange Processes,
Richland, WA
July 1991

* Address: Washington University, St. Louis, MO.

; performed under the auspices of the United States
under Contract No. DE-AC02-76CH00016.

FRACTIONAL ACTIVATION OF ACCUMULATION-MODE PARTICLES IN WARM CONTINENTAL STRATIFORM CLOUDS

N. V. Gillani
N. V. Gillani & Associates Inc., P.O. Box 1141, St. Louis, MO 63043

P.H. Daum and S.E. Schwartz
Environmental Chemistry Division, Brookhaven National Laboratory, Upton, NY, USA

W.R. Leaitch, J.W. Strapp and G.A. Isaac
Cloud Physics Research Division, Atmospheric Environment Service, Downsview, ONT, Canada

ABSTRACT

The degree of activation of accumulation-mode particles (AMP) in clouds has been studied using continuous (1 second average) aircraft measurements of the number concentrations of cloud droplets (N_{cd}, 2 to 35 μm diameter) and of unactivated AMP (N_{amp}, 0.17 to 2.07 μm diameter) in cloud interstitial air. The magnitude and spatial variation of the activated fraction (F) of all measured particles (defined as $F \equiv N_{cd}/N_{tot}$, where $N_{tot}=N_{cd}+N_{amp}$) are investigated, based on measurements made during ten aircraft flights in non-precipitating warm continental stratiform clouds near Syracuse NY in the fall of 1984. Based on instantaneous observations throughout the clouds, the spatial distribution of F was found to be quite non-uniform. In general, F was low in cloud edges and where total particle loading was high and/or cloud convective activity was low. In the interior of clouds, the value of F exceeded 0.9 for 36% of the data, but was below 0.6 for 28%. Factors influencing F the most were the total particle loading (N_{tot}) and the thermal stability of the cloud layer. The dependence of F on N_{tot} in cloud interior was characterized by two distinct regimes. For $N_{tot} < 600$ cm^{-3}, F was generally close to unity and relatively insensitive to N_{tot}. For $N_{tot} > 800$ cm^{-3}, F tended to decrease with increasing N_{tot}. This decrease was greatest in a stable stratus deck embedded in a warm moist airmass. The results suggest that, in warm continental stratiform clouds, the process of particle activation becomes nonlinear and self-limiting at high particle loading. The degree of this nonlinearity depends on cloud convective activity (thermal instability).

1. INTRODUCTION

The concentration, size distribution and chemical composition of cloud droplets influence many important cloud properties (e.g., radiative and chemical) which, in turn, govern the role of clouds in a number of important atmospheric processes (e.g., the natural water cycle, global energetics, atmospheric chemistry, and pollutant redistribution and deposition). These droplets form exclusively on pre-existing aerosol particles (cloud condensation nuclei, CCN). Therefore, the nature of the precursor CCN population in pre-cloud air and the efficiency of CCN activation in clouds are crucial determinants of cloud properties. The recent recognition of the important impact of anthropogenic atmospheric aerosols on cloud optics, and of its significance to the issue of global warming (Twomey et al, 1984), has reaffirmed the importance of the aerosol-cloud link. This aerosol-cloud link is complex and its

To be published in *Precipitation Scavenging and Atmosphere-Surface Exchange*, S. E. Schwartz and W. G. N. Slinn, Coords. Hemisphere, Washington DC., 1992.

characterization based on field measurements is meager. In this paper, we examine the fractional activation of the main mass-bearing portion of atmospheric aerosols (viz., the accumulation-mode particles, AMP) in non-precipitating warm continental stratiform clouds based on in-situ aircraft measurements. Globally, stratiform clouds (stratus and strato-cumulus) are the most widespread; they are also the low clouds exposed to higher concentrations of anthropogenic CCN. Such exposure is greatest over the continents. The difference between continental and marine stratiform clouds is believed to be due mainly to the distinct difference in the particle loading of the two cloud environments (Squires, 1958).

Previous observational studies of nucleation scavenging have focussed on estimation of the scavenged *mass* based on measured concentrations of sulfate and nitrate in air and cloud-water samples (e.g., Scott and Laulainen, 1979; Daum et al, 1984, 1987; Hegg et al, 1984; Hegg and Hobbs, 1986). Typically, such measurements represent averages over 10 to more than 100 km, and the resulting estimates of the scavenging efficiency vary widely and contain large uncertainties (Gillani et al, Unpubl. III). In one study, continuous measurements of particulate sulfur and particle light scattering coefficient (surrogate for particle mass) were used (ten Brink, et al, 1987). Other aerosol scavenging studies have utilized continuous data of particle *number* concentrations either directly to estimate number scavenging (Leaitch et al, 1986; Daum et al, 1987) or indirectly to estimate volume scavenging (Leaitch et al, 1983; Hegg et al, 1984). While the results of the above studies vary widely, the overall conclusion appears to be that nucleation scavenging of sulfate and nitrate mass by clouds is an efficient process.

The activation of CCN in clouds depends on the water supersaturation (S) attained in the cloud updraft under the action of two competing effects: the release of water vapor by cooling in the rising air mass (positively related to updraft speed, w), and the consumption of it by condensation on the CCN (positively related to CCN concentration and hygroscopic nature). The sensitivity of S to CCN concentration is likely to be particularly high in continental stratiform clouds because of the combination of relatively high particle loading and relatively low w. The primary objective here is to determine the dependence of the degree of activation of AMP on total particle loading in warm continental stratiform clouds. The analysis is based on continuous measurements of AMP (> 0.17 μm dia.) and CD (2 to 35 μm dia.) in all parts of clouds. Thus, near-adiabatic as well as non-adiabatic effects are included in the data and the analysis.

2. EXPERIMENTAL

The aircraft measurements reported here were made near Syracuse, NY during Fall 1984 by the Canadian Institute for Aerospace Research Twin Otter aircraft, instrumented for cloud physics and chemistry measurements by the Atmospheric Environment Service of Canada and Brookhaven National Laboratory (for details, see Leaitch et al, 1986; Isaac and Daum, 1987).

Continuous measurements of particle concentrations (at 1 s and ~60 m resolution) in the Twin Otter were made with Particle Measuring Systems probes as follows : PMS-ASASP-100X for aerosols in 15 size classes ranging between 0.17 and 2.07 μm; PMS-FSSP-100 for cloud droplets in 15 size classes ranging between 2 and 35 μm ; PMS -2D-C and -P Imaging Probes, respectively, for cloud drops (25 to 800 μm) and precipitation drops (200 to 6400 μm). Here, the FSSP and ASASP measurements refer to concentrations of the CD and the AMP, respectively. The FSSP data have been corrected for probe dead time and coincidence errors as described by Baumgardner et al (1985). The ASASP in-cloud measurements can include some partially or fully evaporated cloud droplets and cause a slight upward bias. This error, however, is not large enough to have a significant influence on our conclusions. The Twin Otter nose boom vane system provided a measure of vertical gust velocity, but not of the long-wave mean updraft speed, w.

The Twin Otter flew 24 missions between Oct 17 and Nov 14. Ten were selected for analysis based on two criteria: substantial cloud encounter and minimal, if any, precipitation. A total of more than 24 hours of flight data were logged on these days at 1 s resolution. Of these, about 11% were excluded because of possible precipitation (based on flight logs and the 2D-P probe data). Of the remaining records, about 30% were in-cloud (liquid water content, LWC > 0.01 g m^{-3}) and the rest were in clear air. In addition, a total of 48 cloud-water (CW) samples ranging upto 20 minutes in duration were also collected and subsequently analysed for cloud water chemical composition.

Flight times for the selected missions and summaries of the prevailing cloud and meteorological conditions are given in Table 1. The first five of the ten missions were characterized by distinctly warmer cloud conditions. The sampled air masses on three of these days (Oct 18, Oct 26 and Nov 5) were of southerly origin: aerosol number concentration in the air below the clouds, as well as the concentration of ionic species in cloud water (normalized for liquid water content), were highest on these days; the low cloud base in each case indicates the presence of stratus clouds (St). The air masses on the other seven days were of northerly origin, and were substantially cooler and cleaner (lower concentrations of aerosol number and cloud water species mass); with one exception (12 Nov), the cloud bases were significantly higher (St-Cu) on these days.

TABLE 1. Overview of Meteorological and Cloud Conditions

DATE (Twin Otter Flights)	LOCAL TIME	Airmass Origin	Below-cloud N (amp) (cm^{-3})	CLOUD Base - Top (m MSL)	CLOUD Temp (°C)	CLOUD pH
18 Oct	1030 - 1300	SW	1500	400 - 2000	8 - 11	3.2 - 3.4
26 Oct	0930 - 1200	SSW	1500	200 - 2000	4 - 11	3.6 - 4.0
29 Oct	1100 - 1700	WNW	100	2400 - 2800	2 - 5	3.8 - 4.4
31 Oct	1130 - 1400	WNW	900	1000 - 1500?	-1 - 2	3.7 - 4.1
5 Nov	0900 - 1130	SW	1200	200 - 1600	2 - 7	3.7 - 4.4
6 Nov	1130 - 1500	NW	250	1400 - 2400	-14 - -8	3.9 - 4.2
9 Nov	1000 - 1600	WNW	400	1200 - 3600	-8 - 6	4.1 - 4.5
10 Nov	0930 - 1130	NW	100	1600 - 4400	-10 - 6	4.2 - 4.7
12 Nov	1130 - 1430	NNE	500	250 - 3200	-13 - 0	3.4 - 4.2
13 Nov	1200 - 1530	NE	100	700 - 1800	-10 - -5	4.5 - 5.5
FOOTNOTES:		1	2	3		

1. Based on 850 mb 4-day back-trajectory arriving over Syracuse at 18Z
2. Based on the ASASP data of the Twin Otter
3. Local ground elevation is typically 150 - 200 m

3. RESULTS AND DISCUSSION

Aerosol activation occurs when the local supersaturation (S) in a cloud exceeds the critical supersaturation (S_c) required for activation. S_c depends primarily on aerosol size and chemical composition, particularly the size. S, on the other hand, depends principally on updraft speed ($\bar{w}$), the CCN number concentration, and in non-adiabatic parts of the cloud, also on effects related to entrainment and mixing. In this paper, the focus is on the dependence of AMP activation on particle concentration. In a similar study, Leaitch et al (1986) focussed attention on activation *in the adiabatic core near cloud base* by comparing cloud droplet concentration (N_{cd}) there with the precursor AMP concentration (N_{amp}) below cloud base (hence, a Lagrangian approach). Our interest, however, is in examining activation *in all parts of the cloud*. In layered (stratiform) clouds, the vertical structure is generally complex, and it is not always possible to determine with confidence what the pre-cloud precursor particle concentration is corresponding to N_{cd} at any arbitrary cloud location. We have, therefore, adopted an Eulerian approach in which we determine the degree of particle activation locally in all parts of the cloud, without any explicit *a priori* association of activation with pre-cloud conditions. We do this by *local* partitioning of cloud particles into activated droplets and unactivated AMP through a focus on the activated fraction (F) defined as

$$F \equiv \frac{N_{cd}}{N_{tot}} = \frac{N_{cd}}{(N_{cd}+N_{amp})} \quad . \tag{1}$$

This definition has several features which make it useful and robust. F is based only on continuously-measured local variables. Thus, it describes the *local and instantaneous* condition of particle activation. Its evaluation does not require knowledge of conditions below cloud base or in any other part of the cloud surroundings. Each value of F reflects not only the initial activation process, but also the subsequent history of the sampled particle population. Collectively, the values of F describe adiabatic as well as nonadiabatic conditions. The definition is not strictly limited to activation of the AMP size range of dry CCN, but includes also the involvement of those smaller particles which, in the cloud, have either grown into the AMP size range without becoming activated (hence part of N_{amp}), or even those which have become activated (hence part of N_{cd}). In the latter case, N_{cd} may exceed the AMP concentration below the cloud base; nevertheless, by definition, the local value of F cannot exceed unity anywhere in the cloud.

3.1 The Magnitude of F and Its Variation with N_{tot}

N_{tot} should be a conservative property within an adiabatic updraft, and related to the aerosol loading below cloud base in such an updraft. It is also the total measured particle loading in the cloud. In stratiform clouds, in particular, it is approximately constant inside and outside clouds at the same height (Isaac et al, 1990). Therefore, it is a logical independent variable representing airmass particle loading. The F-values for all in-cloud observations of the ten flights are plotted in Figure 1 as a function of N_{tot}; a subset of the same data, averaged over the durations of the 48 CW samples, are plotted in Figure 2 segregated by magnitude of F (low, $F < 0.4$; intermediate, $0.4 < F < 0.8$; high, $F > 0.8$) and day. The dominant characteristics of the magnitude and variations of F based on these figures and Table 1 are highlighted below :

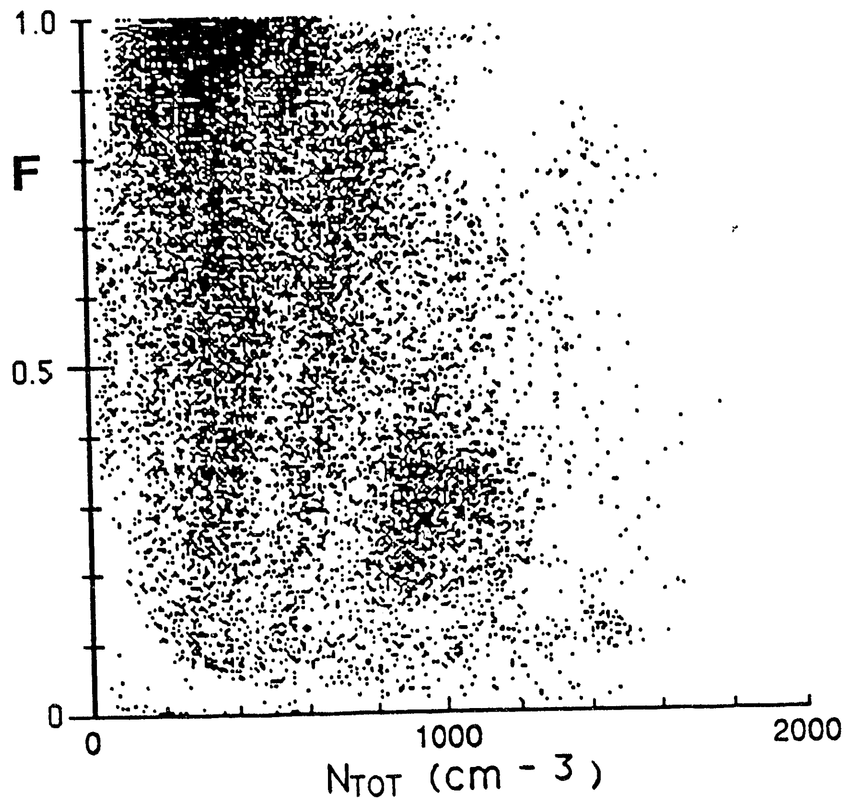


FIGURE 1. F - Data of the Twin Otter for all 10 Days
(~23200 points)

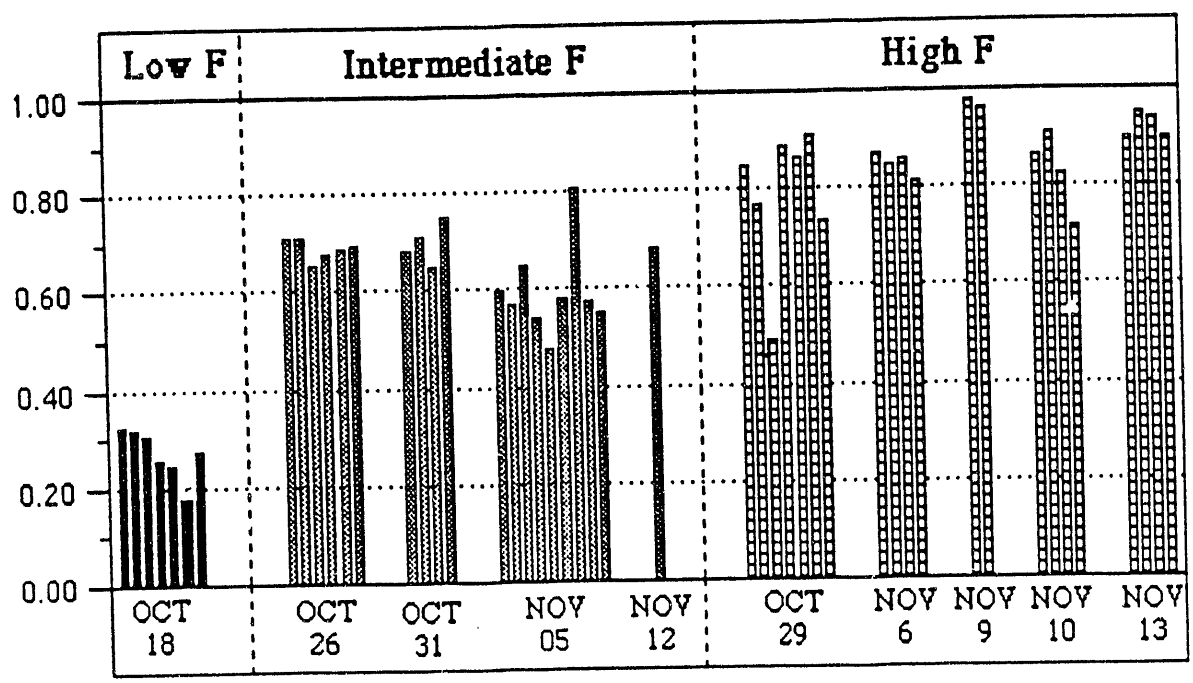


FIGURE 2. Mean Magnitudes of F for Cloud-water Sample Times

1. There is a large cluster of high F values for $N_{tot} \leq 600\ cm^{-3}$, indicating nearly total activation of the AMP and possibly also of some smaller particles (Fig. 1); for any given N_{tot}, however, a wide range of lower F values are also attained.

2. High values of F are generally not attained for $N_{tot} \geq 800\ cm^{-3}$ (Fig. 1); in this regime, values of F appear to decrease with increasing N_{tot}.

3. The clouds on all *five high-F days* (Fig. 2) were St-Cu (in airmasses of northerly origin) with relatively high base heights (Table 1) and neutral-to-unstable lapse rates (Gillani et al, Unpubl I). Below-cloud aerosol concentrations were under 400 cm^{-3}, and cloud-water pH values exceeded 3.8 (Table 1).

4. The clouds on four of *the other five lower-F days* (Fig. 2) included low St with stable-to-neutral lapse rates. Below-cloud aerosol concentrations were much higher (up to 1500 cm^{-3}), and cloud-water pH significantly lower (as low as 3.2), particularly on the three days with southerly airmasses (Oct 18, 26, and Nov 5).

It is instructive to examine the data of Figure 1 in various disaggregated forms. Figure 2 represents one such disaggregation in which the data are divided into groups of days with different average magnitudes of F (over 5 to 20 minute durations). Figure 3 shows a similar breakdown, but here the data are unaveraged (beyond 1 s). The figure shows, first of all, different characteristic patterns of the dependence of F on N_{tot} for high-F versus low-F days. The high-F days (Fig. 3a) all exhibit a predominant cluster ("head") of points with F near unity, and a "tail" made up of much fewer points extending down to F~0 for similar N_{tot}. The N_{tot} values on these days generally remain under 600 cm^{-3}. In contrast, most N_{tot} values on the low-F day (Fig. 3b) exceed 800 cm^{-3}, and there is a sharp drop-off in F with increasing N_{tot}; the dominant feature here is the cluster of points with F in the range 0.2 to 0.4 for $N_{tot} \sim 800$ to 1200 cm^{-3}. A similar drop-off was inferred by Leaitch et al (1986) for $N_{amp} > 750\ cm^{-3}$ at cloud base. Such limited activation was also observed at high particle loading by Fitzgerald and Spyers-Duran (1973) in small Cu and St-Cu within the urban plume of St. Louis. The data of Oct 31 (Fig. 3c) have been isolated from those of the other three intermediate-F days (Fig. 3d) because they clearly show the transition between features of the low-N behavior (head and tail for $N_{tot} < 800\ cm^{-3}$) and those of the high-N behavior (drop-off in F as N_{tot} increases beyond 800 cm^{-3}). The intermediate-F status assigned to this day in Figure 2 is a result of averaging across the two different regimes; a large number of individual F values are above 0.8. By contrast, the intermediate-F status of the other three days (Oct 26, Nov 5, 12; Fig. 3d) is due to the fact that the bulk of the F values fall in the intermediate range within a single N-regime ($N_{tot} \leq 800\ cm^{-3}$). The corresponding picture (Fig. 3d) shows mostly the "tail" (without the "head") of the low-N behavior, and a rather small number of points for $N_{tot} > 800\ cm^{-3}$ which exhibit the corresponding trend of decreasing F with increasing N_{tot}.

3.2 Spatial Variation of F: The Edge Effect

In cloud edges, particularly near cloud top and sides, S (and hence F) can be depressed as a result of entrainment of drier air from outside the cloud. This effect can be quite significant in stratiform clouds owing to their long lifetimes. The 1-D adiabatic assumption often invoked in cloud studies is thus useful in updraft cores near cloud base, but breaks down near cloud edges and in the upper portions of clouds where significant entrainment and mixing effects may prevail. The high-F cluster at low N_{tot} (Fig. 3a) is consistent with 1-D adiabatic theory near cloud base (for example, Twomey 1959; Twomey and Warner 1967; Jensen and Charlson 1984;

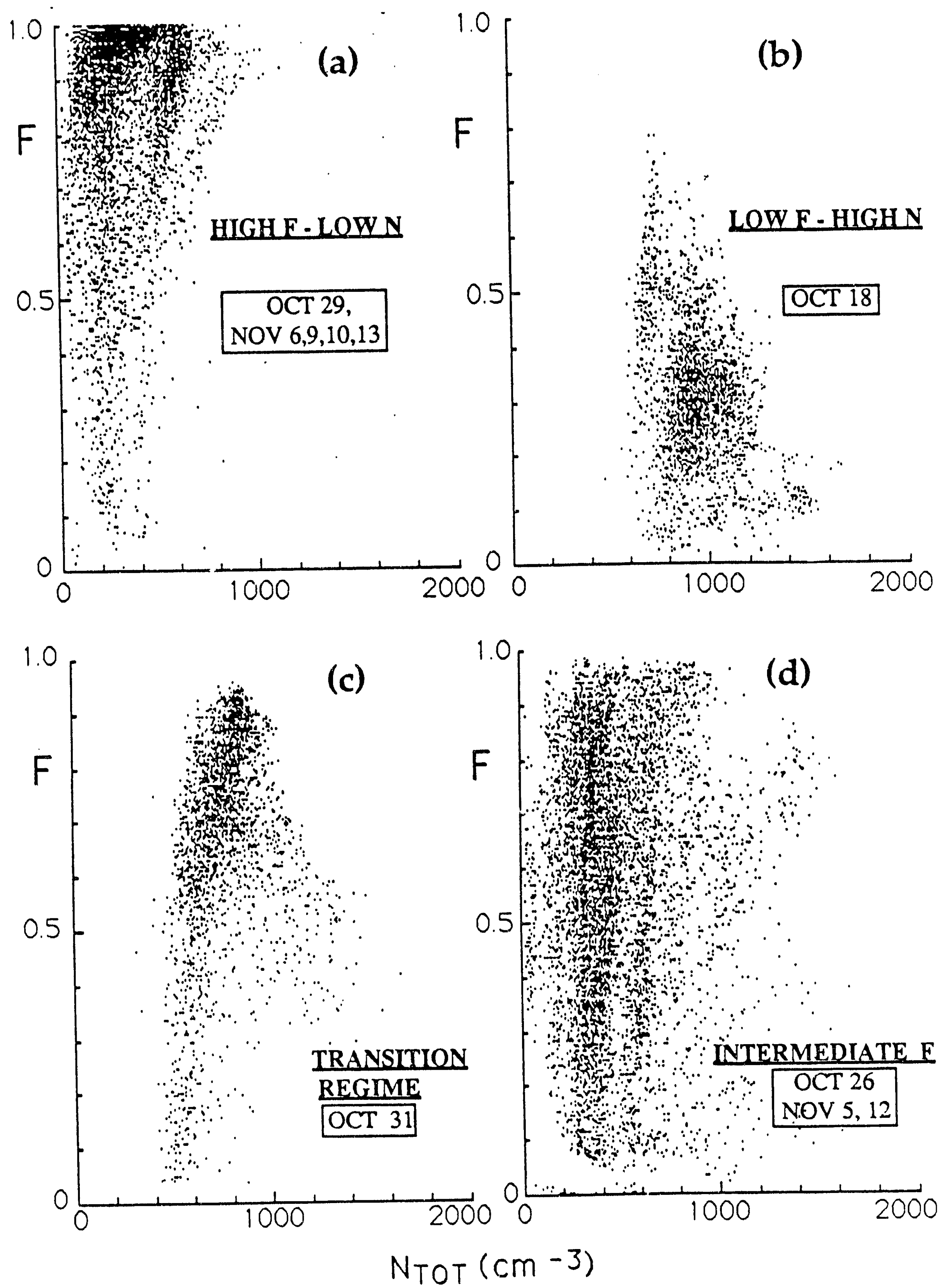


FIGURE 3. The Variation of F with N_{tot}
Classification by Mean Magnitude of F

Leaitch et al, 1986), which predicts a quasi-linear relationship between the concentrations of CD and of the parent population of AMP under conditions of low N_{tot} (slope ~ F ≈ constant). The near-unity value of the slope, indicating almost total activation of aerosol mass, is also consistent with this theory (Jensen and Charlson 1984). The adiabatic assumption, however, becomes questionable for points in the "tail", because many of them have entrainment effects present. One example illustrating this is shown in Figure 4 for the data of Nov 9. The plots in Figure 4a show F nearly constant (~1) during a horizontal traverse near cloud base through a more or less solid cloud deck (as indicated by the LWC data). The N_{tot} values were mostly under 600 cm^{-3} and most of these data correspond to the "head" (see right panel) and represent quasi-adiabatic conditions. In contrast, the plots in Figure 4b show conditions resulting in the "tail" (see right panel). Most of these data correspond to cloud edges and are likely to include non-adiabatic effects. A different type of situation is shown in Figure 5 (Oct 18) in which non-adiabatic effects may be present even in the interior of an aged stable stratus deck, and which may be at least partially responsible for the depression of F. In this case, the presence of non-adiabatic effects is inferred from droplet size spectra, which show a multi-modal structure uncharacteristic of adiabatic condensation (Gillani et al, Unpubl. II). The low F in Figure 5 is due to low S resulting from a combination of factors: high N_{tot}, absence of a significant updraft, stable thermal stratification, coalescence, and entrainment effects (Gillani et al, Unpubl. IV). Another example of non-adiabatic and edge effects present in the "tail" is shown in Figure 6 for the data of October 26 (see also Fig. 3d). The vertical profiles of cloud water (LWC) and temperature (Fig. 6a) show extensive stratification. The multiple cloud tops and many edges are conducive to entrainment and non-adiabatic effects, and the multiple temperature inversions inhibit convection and updrafts. Thermal instability, on the other hand, appears to enhance the degree of activation (Gillani et al, Unpubl. I). Even though N_{tot} is mostly under 600 cm^{-3}, values of F show a conspicuous absence of the "head" and exhibit mostly only the "tail" (Fig. 6b).

The contrast in the degree of activation in cloud interior versus cloud edges is further illustrated in Figure 7 which shows log-log plots of F versus N_{tot}. Figure 7a shows all in-cloud data points (~23200); Figure 7b shows a subset of these data, called "cloud interior" (~17400 points). Excluded in Figure 7b are the data in cloud "edges" defined as all points for which LWC < 0.03 g m^{-3}, as well as all points sampled within 3 s from such points (aircraft speed ~ 60 m s^{-1}). In Figs. 7 a and b, the locus of points of maximum F over the full range of N_{tot} (~ 20 to 1700) defines a maximum-F curve (MFC). The MFC is nearly flat (F≈1) for $N_{tot} < N_{tot}^*$ (~ 600 - 800 cm^{-3}), and drops off at higher N_{tot}. The points on this curve represent conditions in which factors other than N_{tot} contributing to S and F (e.g., updraft speed, cloud con-vective activity and adiabaticity, etc.) are optimum. Points below the MFC (the "tails") are due to variations in these other factors. In Figure 7b, many of the points in these "tails" for N_{tot} < 800 cm^{-3} belong to the intermediate-F days (Fig. 3d), and are similar to those of October 26 (Fig.6b) which include non-adiabatic effects related to layering of the clouds (more surface area), as well as complex vertical thermal structure. We suggest that the MFC mimics some important features of the well-known empirical form of CCN activation spectra ($N_{act} = C. S^k$), with F representing the degree of activation and increasing-N serving qualitatively as a surrogate for decreasing-S. The inverse relationship between S and N_{tot} is as expected (Twomey 1959). The surrogate activation spectrum represented by the MFC (of the form $F \sim N_{tot}^{-\alpha}$) exhibits two distinct regimes separated by the transition region characterized by N_{tot}^*. A similar two-regime dichotomy of CCN spectra was also observed by Alofs and Liu (1981) for $S < S^*$ and $S > S^*$, with S^* consistently near 0.05%. One regime (the flat part: $N_{tot} < N_{tot}^*$, $S > S^*$) is characterized by a

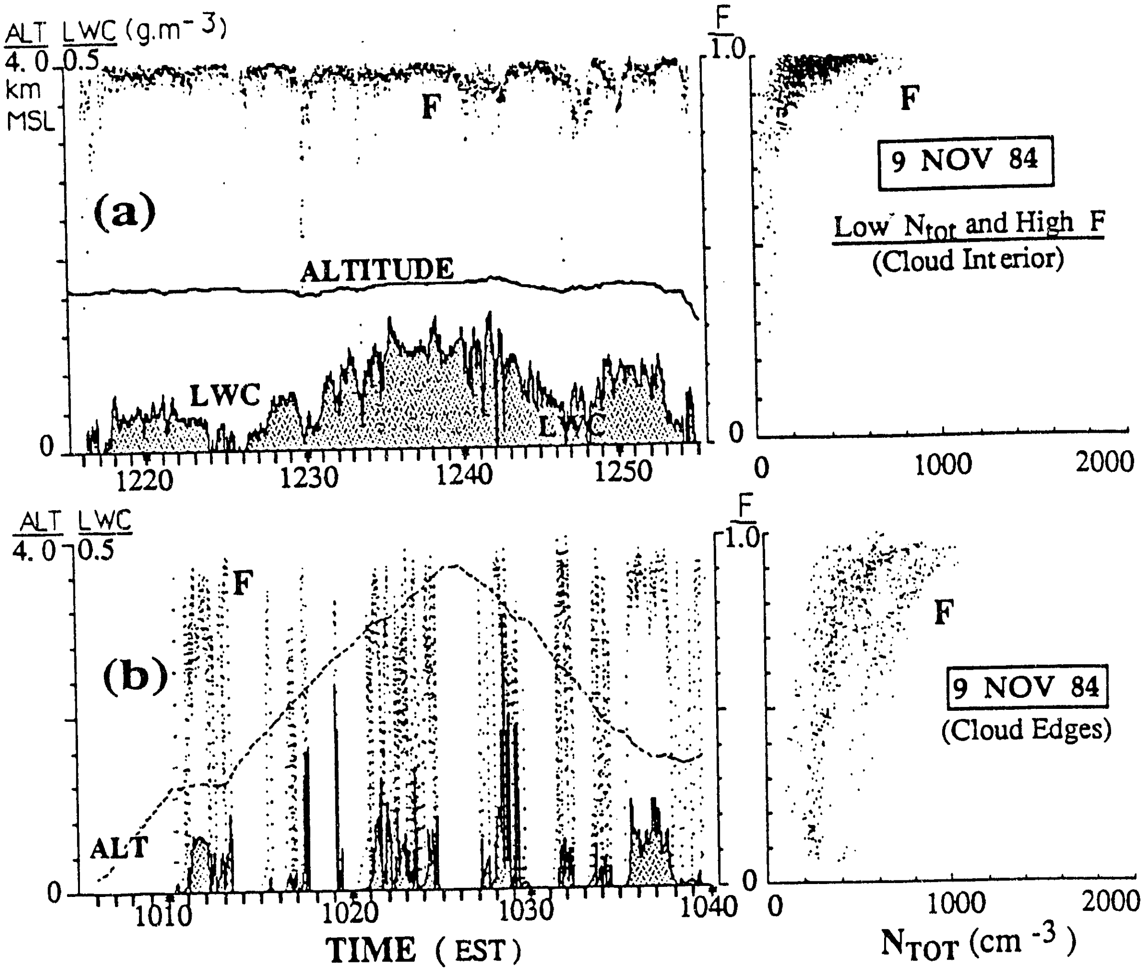


FIGURE 4. F in Cloud Interior and Cloud Edges for Low N_{TOT}

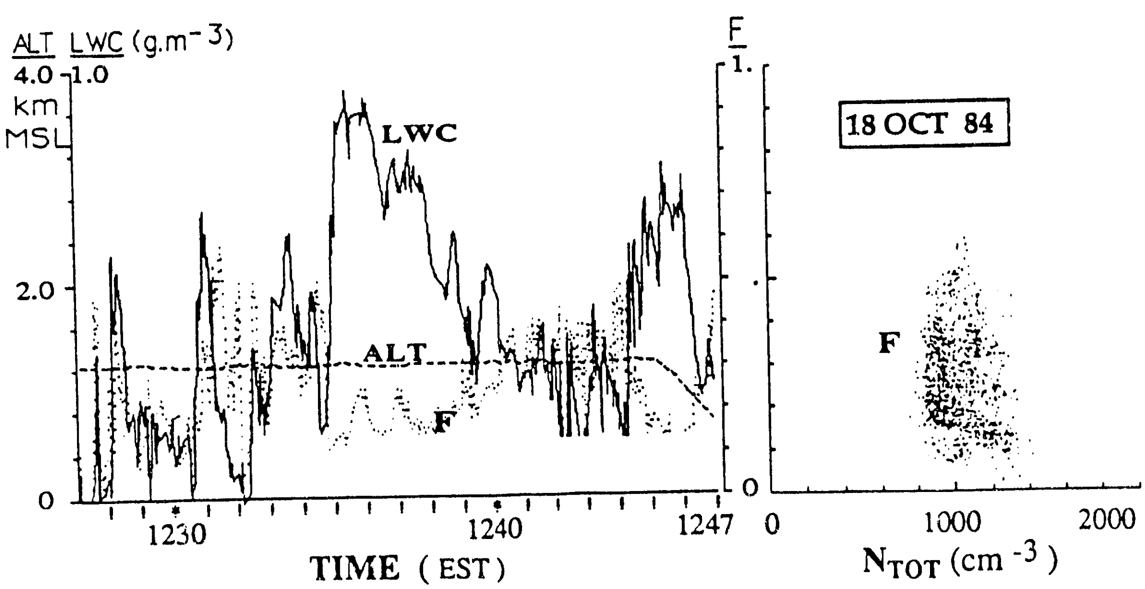


FIGURE 5. Low F in the Interior of a Stable Stratus Cloud for High N_{TOT}

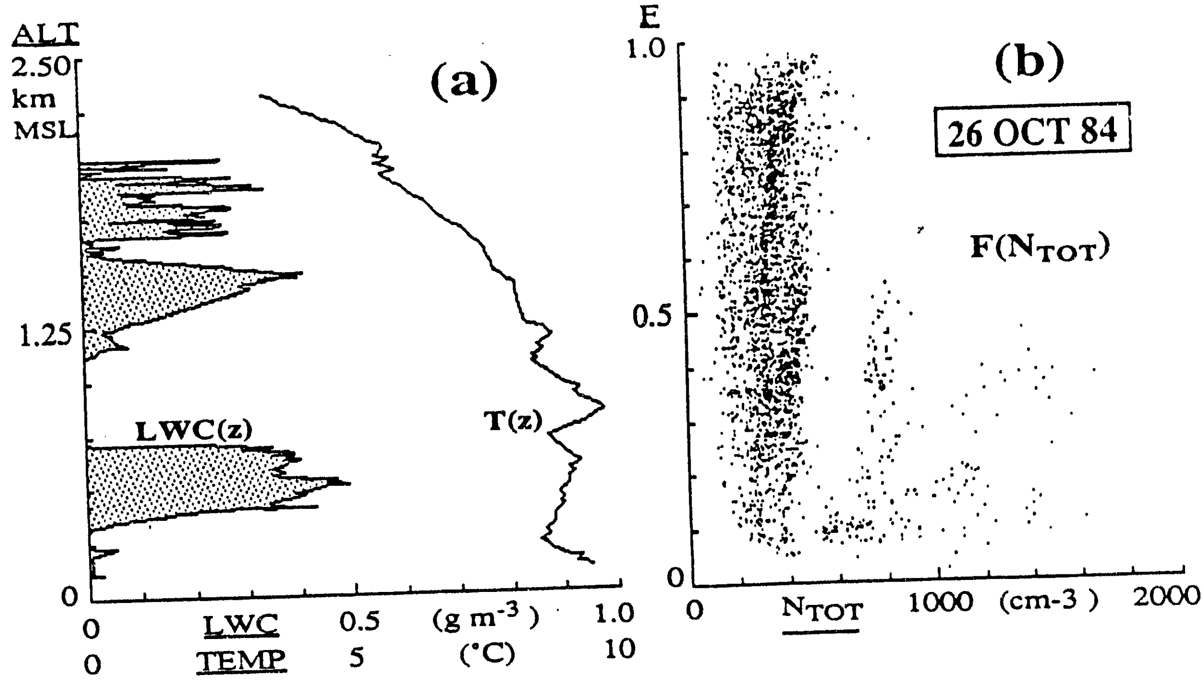


FIGURE 6. Particle Activation (F) in a Complex Vertical Cloud Structure

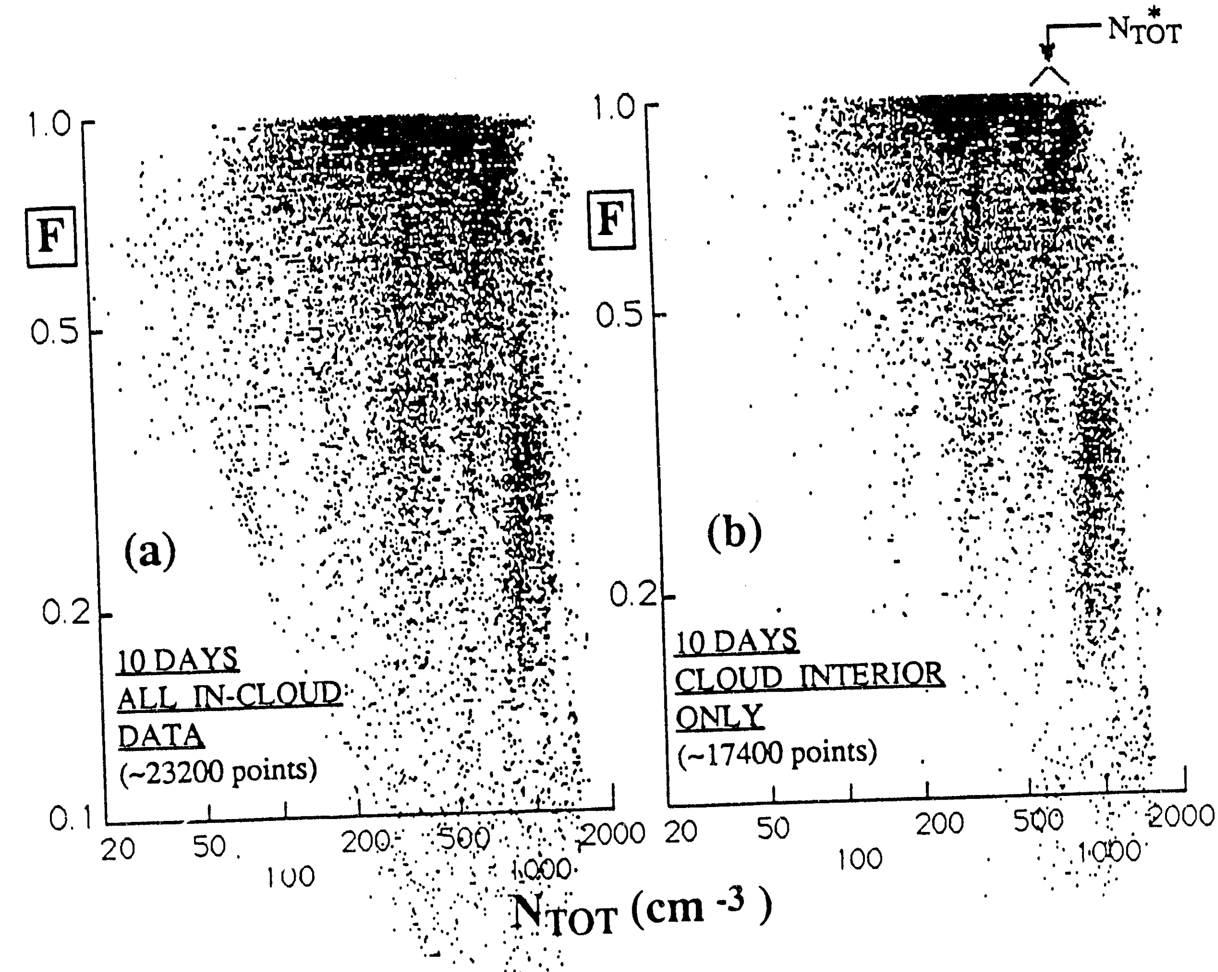


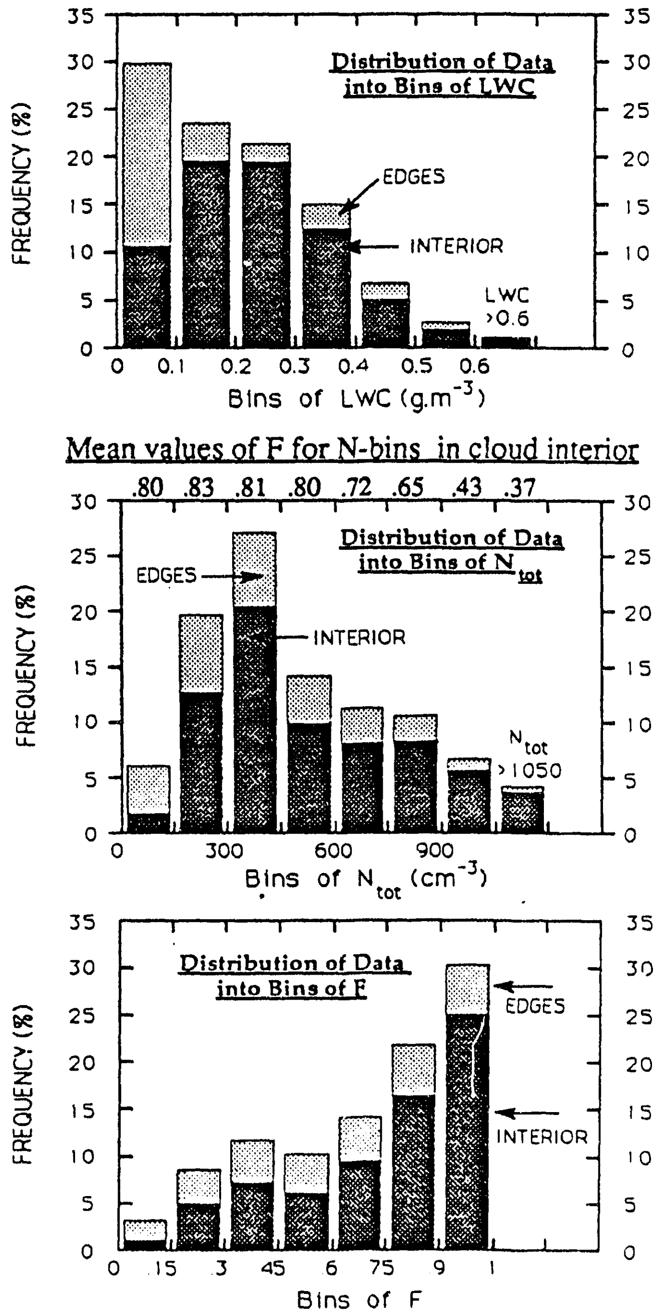
FIGURE 7. Degree of Activation : The Edge Effect

high degree of activation of a relatively sparse AMP population. In this case, the demand for water by the low N_{tot} is easily met by a generous supply (relatively high S). Thus, the activation process is rather insensitive to increase in N_{tot} (or decrease in S) as long as $N_{tot} < N_{tot}^*$. The other regime (the steep part: $N_{tot} > N_{tot}^*$, $S < S^*$) is characterized by activation which is quite sensitive to N_{tot} (and S), because of a delicate balance between high demand for the condensate (by the high N_{tot}) and a limited supply (low S). Based on the adiabatic theory of Twomey (1959), the drop-off rate 'α' of F can be shown to be related to the exponent 'k' of the CCN spectra by the relation $\alpha = k/(k+2)$. For three days with high N_{tot} data (Oct 18, 31, and Nov 5), Gillani et al (Unpubl I) show α to vary between about 0.25 and 1, corresponding to $k > 2/3$. For the low N_{tot} data, in general, $\alpha < 0.25$ (i.e., $k < 2/3$). 'α' and 'k' are measures of the sensitivity of the activation process to N_{tot} and S, respectively. Based on the dependence of S_c on CCN size and composition, as given in Alofs et al (1989), nearly total activation of continental AMP ($D > 0.17$ mm) may be expected at $S \approx 0.1\%$. It may be concluded that, in the Syracuse study, S was probably somewhat higher than 0.1% under the high-F conditions, and possibly less than 0.05% in the stable clouds in which N_{tot} exceeded 800 cm^{-3}. S is, of course, a difficult quantity to measure or estimate, and the corresponding uncertainty is high.

FIGURE 8.

Classification of Cloud Data by F, LWC, and N

(% frequency based on about 23200 in-cloud data points)



3.3 Frequency Distribution of the Cloud Data With Respect to LWC, N_{tot} and F

Figure 8 illustrates three histograms of the in-cloud data. The lighter shaded bars represent all in-cloud data (~23200 observations). The superimposed darker shaded bars represent the "interior" data of all ten days (i.e., at least 3 s from LWC < 0.03 g m^{-3}). The "interior" data constitute about 75% of the total. In the "interior", the liquid water content is predominantly (53%) between 0.1 and 0.3 g m^{-3}. 60% of the N_{tot} values are between 150 and 600 cm^{-3}, and in 23% of the data, N_{tot} > 750 cm^{-3}. F exceeds 0.9 for 35% of the interior data, but is under 0.6 for 28% of the data.

4. CONCLUSION

The activation efficiency function (F) is viewed as a useful descriptor of the local state of AMP activation in clouds. Its determination is based directly and straightforwardly on the measured values of N_{amp} (ASASP) and N_{cd} (FSSP). The use of F permits quantitative and detailed examination of an important property of the aerosol-cloud link. Its application extends over adiabatic as well as nonadiabatic portions of the cloud.

The vast majority of published data of CCN activation spectra are in the range $S > 0.1\%$. The bulk of CCN mass (in the accumulation and higher modes), however, is activated at $S < 0.1\%$, and it is in this latter realm that the activation process is most sensitive to S (particularly through N_{tot} and cloud convective activity). For the continental stratiform clouds we have studied, this high-sensitivity realm appears to correspond to $N_{tot} > 800\ cm^{-3}$. Similar studies need to be performed in more convective clouds, and in maritime clouds, so that a deeper and more generalized understanding of cloud formation and microphysics can be gained. Conditions least conducive to high F in stratiform clouds were found in warm, moist and stable airmasses with high particle loading. Specifically, when N_{tot} exceeded about 800 cm^{-3} in these clouds, the supersaturation conditions were apparently such that only a limited fraction of the AMP could be activated. For higher N_{tot}, the activation process became increasingly self-limiting. Such decreased efficiency of nucleation scavenging from air with high particle loading in industrialized regions has important implications for precipitation formation and acidification, for long range transport of the unactivated particles, and for cloud radiative properties and global energetics.

Acknowledgement. This work was conducted under Contract No. DE-AC02-76CH00016 with the U.S. Department of Energy, and under Contract No. RP2023-1 with the Electric Power Research Institute. We also gratefully acknowledge the contributions of the Canadian Institute for Aerospace Research.

5. REFERENCES

Alofs D.J. and Liu T. (1981) Atmospheric measurements of CCN in the supersaturation range 0.013 - 0.681%. *J. Atmos. Sci.* **38**, 2772-2778.

Alofs D.J., Hagen D. E. and Trueblood M.B. (1989) Measured spectra of the hygroscopic fraction of atmospheric aerosol particles. *J. Appl. Met.* **28**, 126-136.

Baumgardner D., Strapp J. W. and Dye J. E. (1985) Evolution of the forward scattering spectrometer probe, part II. Corrections for coincidence and dead time errors. *J Atmos. Oceanic Technol.* **2**, 626-632.

Daum P.H., Kelly T., Schwartz S.E. and Newman L.(1984) Measurements of the chemical composition of stratiform clouds. *Atmos. Environ.* **18**, 2671-2684.

Daum P.H. et al, (1987) Chemistry and physics of a winter stratus cloud layer : A case study. *J. Geophys. Res.* **92**, 8426-8436.

Fitzgerald J.W. and Spyers-Duran P.A. (1973) Changes in cloud nucleus concentration and cloud droplet size distribution associated with pollution from St. Louis. *J. Appl. Met.* **12**, 511-516.

Gillani N.V. et al, (Unpubl I,II,III,IV. In preparation for journal publication) Field observations in warm continental stratiform clouds :
I. Fractional activation of accumulation-mode particles.
II. The degree of activation and cloud microstructure.
III. Partitioning of sulfate and nitrate mass between air and water in clouds.
IV. Formation of warm precipitation - A case study.

Hegg D.A., Hobbs P.V. and Radke L. F. (1984) Measurements of the scavenging of sulfate and nitrate in clouds. *Atmos. Environ.* **18**, 1939-1946.

Hegg D.A. and Hobbs P.V. (1986) Sulfate and nitrate chemistry in cumuliform clouds. *Atmos. Environ.* **20**, 901-909.

Isaac G.A. and Daum P.H. (1987) A winter study of air, cloud and precipitation chemistry in Ontario, Canada. *Atmos. Environ.* **21**, 1587-1600.

Isaac G.A., Leaitch W.R. and Strapp J.W. (1990) The vertical distribution of aerosols and related compounds in air and cloudwater. *Atmos. Environ.* **24A**, 3033-3046.

Jensen J.B. and Charlson R.J. (1984) On the efficiency of nucleation scavenging. *Tellus* **36B**, 367-375.

Junge C.E. (1963) ***Air Chemistry and Radioactivity.*** Acad Press, NY.

Leaitch W.R., Strapp J.W., Wiebe H.A. and Isaac G.A. (1983) Measurements of scavenging and transformation of aerosol inside cumulus. In ***Precipitation Scavenging, Dry Deposition and Resuspension*** (eds. Pruppacher H.R., Semonin R.G. and Slinn W.G.N.) **Vol 1**, 53-69; Elsevier , NY.

Leaitch W.R., Strapp J.W. and Isaac G.A. (1986) Cloud droplet nucleation and cloud scavenging of aerosol sulfate in polluted atmospheres.*Tellus* **38B**,328-344.

Scott B.C. and Laulainen N.S. (1979) On the concentration of sulfate in precipitation. *J. Appl. Met.* **18**, 138-147.

Squires, (1958). The microstructure and colloidal stability of warm clouds: I. The relation between structure and stability. *Tellus* **10**, 256-261.

ten Brink H.M., Schwartz S.E. and Daum P.H. (1987) Efficient scavenging of aerosol by liquid-water clouds. *Atmos. Environ.* **21**, 2035-2052.

Twomey S. (1959) The nuclei of natural cloud formation. Part II : The super-saturation in natural clouds and the variation of cloud droplet concentration. *Geofis. pura e appl.* **43**, 243-249.

Twomey S. and Warner J. (1967) Comparison of measurements of cloud droplets and cloud nuclei. *J. Atmos. Sci.* **24**, 702-703.

Twomey S.A., Piepgrass M. and Wolfe T. L. (1984) An assessment of the impact of pollution on global cloud albedo. *Tellus* **36B**, 356-366.

6. DISCUSSION

A. L. WILLIAMS. Even for these clouds of low peak supersaturation that you have examined, the value of F tends to be close to unity for most cases. Doesn't this imply that CCN of sizes less than AMP are also being activated?

N. V. GILLANI. The possibility is certainly there, and we have alluded to it briefly in the paper. We have seen occasional indications that particles finer than 0.17 mm dry diameter may have been activated (and contributed to N_{cd}), or at least may have grown into the AMP size range in the cloud air without becoming activated (thereby contributing to N_{amp}). In either case, such particles get counted either as N_{cd} or N_{amp}, and hence are accounted for in the local value of F. Assuming similar composition of the CCN of different sizes, activation of the finer particles would imply complete or nearly complete activation of the AMP. In such cases, therefore, F must be close to unity. This appears to happen mostly when the cloud thermal structure is relatively unstable and $N_{tot} < 600\ cm^{-3}$. Presumably, such cases correspond to $S \gtrsim 0.1\%$.

W. G. N. SLINN. How do your results compare with Junge's estimates of the nucleation scavenging efficiency?

N. V. GILLANI. About 30 years ago, Junge (1963) gave the following estimates of the nucleation scavenging efficiency: 0.5 for continental aerosol of high concentration to 0.8 for continental aerosol of low concentration; and 0.9 to 1.0 for marine aerosol ($N < 200 - 300\ cm^{-3}$). In view of our results, Junge's estimates clearly display considerable foresight. The significance of particle loading as a major influencing factor was clearly well recognized. Our results point out, in addition, the important significance also of cloud convective activity and non-adiabatic effects. Further systematic study of the role of all these important factors, as well as of CCN size and composition, under a broader variety of conditions (continental and marine, stratiform and cumuliform, etc.) will, hopefully, make it possible to develop and incorporate meaningful parameterizations of important micro-physical cloud processes in regional and global models with acceptable and assessible uncertainty.

END

DATE FILMED

12 / 29 / 92