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Aerosol particles in the mexican east pacific part I: processing and vertical redistribution by clouds

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Abstract

Airborne measurements of aerosol particle size distributions were made in the Mexican Intertropical Convergence Zone. The shape of the spectra at cloud base was compared with those at higher altitudes and near cloud boundaries to identify signatures of cloud processed particles. Of 78 cases analyzed, 71% showed enhancement in volume of super-micron particles, 49% had enhanced volume of sub-micron particles, 28% were homogeneous mixtures with boundary layer air, and 24% had super-micron particles removed by precipitation. Almost 100% of the cases with enhanced volume in sub-micron particles also occurred with enhanced super-micron volume. The enhanced volume in super-micron particles is approximately 10 times larger than the sub-micron enhancement. Cloud processed particles in marine air masses had twice as much enhancement of super-micron mass than found in particles processed by clouds formed from continental sources, likely a result of a more efficient coalescence process in clean, maritime clouds. These results are in qualitative agreement with previous observational and theoretical studies that relate enhancements in particle mass to the uptake by cloud droplets of SO_2 and subsequent growth by coalescence.

1. Introduction

Aerosol particles (AP) have a dominant role in the formation of clouds, their optical properties and their lifetimes. The fundamental process by which a cloud droplet forms on an AP, i.e. a cloud condensation nucleus (CCN), is well understood and can be described theoretically and validated with observations if the composition of the particles is known (e.g. Twomey, 1991; Raga and Jonas, 1993a, b). On the other hand, the fate of AP after processing by clouds, i.e. changes in size, concentration and composition, is less well understood. Clouds are a transport mechanism that removes AP from the boundary layer and redistributes them through a variety of dynamical and microphysical mechanisms, e.g. entrainment, mixing and precipitation (Flossmann, 1998). It is

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generally accepted that some fraction of the AP incorporated into cloud particles will eventually appear again as AP but in a chemically or physically altered form. For example, aircraft measurements have found that AP sizes can increase in regions of high relative humidity (RH) near clouds (Baumgardner and Clarke, 1998) and other studies (Saxena et al., 1970; DeFelice and Cheng, 1998; Kaneyasu et al., 2001) found that condensation nuclei (CN) concentrations are often higher in the outflow from evaporating clouds than in the environment further away. The AP mass will change as a result of particle sulfate and SO_2 scavenging (Hegg and Hobbs, 1981, 1982; Flossmann et al., 1985, 1987; Flossmann and Pruppacher, 1988; O'Dowd et al., 2000).

There have been a number of observational studies of AP interactions with clouds (Hegg and Hobbs, 1981, 1983; O'Dowd et al., 2000), but very few in the Pacific Intertropical Convergence Zone (ITCZ) and none in the Mexican East Pacific ITCZ. This region is unique as a result of the variation in AP sources. When the wind direction is westerly the AP are of maritime origin and total number concentrations are less than 500 cm^{-3}. Northerly and easterly winds, however, bring continental, anthropogenic AP from Mexico or Central America and the average total number concentrations are greater than 1000 cm^{-3}. These differences in AP origins have significant consequences on cloud formation and the fate of the AP when they are processed by clouds. This paper, part one of two parts, evaluates airborne measurements of AP and cloud particles in the ITCZ of the Mexican East Pacific and shows how AP properties are affected by cloud processing. Part two will discuss the direct and indirect effect of AP in this region of the tropics.

2. Measurement and analysis methodology

2.1. Research area and sampling strategy

The data used in this study were obtained during flights of the US National Science Foundation C-130 research aircraft during the 2001 East Pacific Investigation of Cli-

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mate (EPIC) project. The program and scientific objectives are described in the EPIC science and implementation plan (Raymond et al., 2004). Nine of the nineteen flights between 1 September and 16 October 2001 were dedicated to AP and cloud studies. These flights were within the region 8°–12° N, 93°–97° W, approximately 800–1000 km from Mexico and Central America in the ITCZ region (Fig. 1). Measurements in and around clouds were made at constant levels starting at 4200 m and then descending to 2500 m, 1000 m, 300 m (cloud base) and 30 m a.s.l. The clouds in the research area consisted of small fields of shallow cumulus and lines of deep convection. The investigation concentrated on these latter clouds that generally developed from the southwest to northeast. The flight trajectories were across and along the cloud lines at each level. The across-the-line patterns normally sampled in the regions of newer growth and the along-the-line flight legs sampled cells in both developing and decaying stages. In the across-the-line flight tracks the aircraft sampled cloud-free air for at least 20 km on each side of the cloud.

2.2. Instrumentation

The physical and optical properties of AP are derived directly from the particle size distribution (PSD), a composite synthesized from the measurements by three optical particle spectrometers: PCASP, FSSP-300 and FSSP-100 that measure in the size ranges 0.1–3 μm, 0.3–20 μm and 2–50 μm, respectively. The concentrations in overlapping size ranges were averaged prior to creating the composite. The concentration of condensation nuclei (CN) was measured with a TSI Model 3760, measurements of Cloud Condensation Nuclei (CCN) were made with a University of Wyoming Static Diffusion Cloud Chamber, and the coefficients of light scattering and absorption were measured with a Radiance Research nephelometer and particle soot absorption photometer (PSAP), respectively. All measurements were averaged into one second intervals except the CCN that takes a measurement every 60 s. The technical specifications of these instruments are listed in Table 1.

The measurement uncertainties associated with the PCASP have been discussed

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by a number of authors (Kim and Boatman, 1990; Strapp et al., 1992) as have those of the FSSP-100 and FSSP-300 (Baumgardner et al., 1985, 1992; Baumgardner and Spowart, 1990). The PSAP measurements were corrected using the scattering coefficient from the nephelometer and an empirical factor (Bond et al., 1999).

The three optical size spectrometers were mounted on the wing pods of the C-130. A heater on the inlet to the PCASP reduces the relative humidity (RH) of the sampled air stream to less than 30% (Strapp et al., 1992); hence, the PCASP measures AP whose water content has been removed. The FSSP-100 and FSSP-300 measure particles at the ambient RH. In order to combine the measurements from the FSSP 100 and 300 with those of the PCASP, the spectra were adjusted to a dry equivalent. This adjustment assumes that the measured particles are deliquesced NaCl with the amount of water mass on the particle determined by the ambient RH (Tang, 1980). The resulting sizes are calculated by removing the predicted water mass.

The CN, CCN, PSAP and nephelometer were mounted in the cabin and sampled from inlets mounted on the aircraft exterior. The CN inlet was mounted on the bottom of the fuselage with a special housing to minimize spurious counting caused by drop breakup on the inlet lips. Examination of the CN measurements when in rain indicated that this housing was ineffective and CN during these periods are not included in the current analysis. The CCN, PSAP and nephelometer shared a common, backward facing inlet to minimize droplet shattering and to allow measurements of interstitial aerosols in cloud. The largest size that can enter this inlet is approximately one micrometer, a threshold that has been established from previous studies (Baumgardner et al., 1991).

2.3. Analysis of cloud processing signatures

Cloud processing of AP changes their size, concentration or composition. These changes are reflected in differences in the shapes of PSDs measured “near cloud” compared with two reference PSDs – those at cloud base and “far from cloud”. The cloud base PSD is used as a reference under the assumption that the majority of cloud

droplets form on CCN that enter through cloud base (Flossmann, 1998). The "far from cloud" PSD is measured at the same altitude as "near cloud" but at a distance from cloud where it is assumed the air has not been mixed with "near cloud" air. In the present study, near-cloud is defined as those regions where the equivalent potential temperature, Θ_e, is at least one degree different than the environmental Θ_e, determined from a vertical profile made in a clear air region far from clouds. The time series in Fig. 2 shows two "near cloud" regions where one side is designated as the "growing" side (69 922–69 929) and the other the "dissipating" side (70 141–70 181). The cloud edge is determined from the droplet concentrations measured by the FSSP-100, whose lower size threshold is approximately 2 μm. When the droplet concentration exceeds 1 cm^{-3} the measurements are defined as "in-cloud". This criterion is based on an evaluation of measurements at an altitude of 30 m where there are no clouds and only super-micron sea salt particles will be measured by the FSSP-100. These super-micron particles are the first to be activated at low supersaturations during cloud formation, hence their average concentration of <1 cm^{-3} sets the lower limit for the cloud droplet concentration when there has been no entrainment and mixing with droplet-free air.

A growing region is distinguished from a dissipating side by the rate at which the Θ_e changes as cloud edge is approached and by its variability. In the example shown in Fig. 2, Θ_e on the growing side increases from the environmental value of 338 to the in cloud value of 344 in ten seconds of flight (approximately 1000 m) but on the other side of the cloud, in the dissipating region, the same Θ_e change occurs over 4000 m.

In the following analysis, we assume that the majority of AP at cloud edges is a mix of cloud processed and ambient air. We also assume that the AP in cloud, either nuclei of cloud droplets or interstitial, have originated at cloud base. Finally, we assume that our technique for removing water mass as a function of ambient RH provides a conservative estimate of the dry diameter such that size changes by deliquescence will not be a factor in the evaluations. In the following analysis, the PSD are presented as volume concentrations ($\mu m^3\, cm^{-3}$) as a function of size. This emphasizes the sizes changes

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by cloud processing and underscores the importance of volume changes since particle mass, fall velocity and heterogeneous chemistry depend on the volume more than on number concentration. Volume changes are also a reflection of mass increases that result from in-cloud aqueous chemistry or droplet coalescence.

Based upon a visual inspection of the measured PSDs, four patterns have been identified that are associated with cloud processed particles: 1) enhanced concentrations of sub-micron AP ($\leq$1 μm), 2) enhanced concentrations of super-micron (>1 μm), 3) removal of super-micron particles and 4) dilution with minimal changes in the shape of the PSD.

Figure 3a illustrates Types 1 and 2, enhancement of sub and super micron particles with respect to the cloud base PSD. The near cloud measurements in this example were made at 1000 m and cloud base is always at 300 m. The concentration of particles less than 0.3 μm in the far cloud measurements are the same as at 300 m but decrease at larger sizes. The near cloud concentrations are greater at all sizes. Concentrations increase with respect to the cloud base AP by four mechanisms: 1) mixing with ambient air whose particle concentration is higher than at cloud base, 2) growth of particles with sizes smaller 0.1 μm, 3) droplet coalescence, or 4) AP scavenging by cloud droplets. Given that the ambient AP concentration shown in Fig. 3a is the same or lower than cloud base, the 1st mechanism does not contribute to enhanced concentration. The minimum size threshold of the PCASP is 0.1 μm; hence, AP with sizes less than 0.1 μm may grow as a result of cloud processing into the range of the PCASP. AP can grow as a result of chemical reactions whereby ambient gases like SO_2 are dissolved in water droplets and aqueous chemistry increases the mass of the original CCN (Hegg et al., 1982, 1983; Flossmann et al., 1987; Flossmann and Pruppacher, 1988; O'Dowd et al., 2000; Alfonso and Raga, 2002). This mechanism will increase the mass of all CCN within cloud droplets after evaporation, as suggested in the near cloud PSD of Fig. 3a whose peak at 0.8 μm is slightly higher than the peak of the cloud base PSD found at 0.5 μm. The AP in a drop formed by coalescence of droplets is the combined mass of the CCN in the original droplets (Flossmann et al., 1987; Alfonso and Raga, 2002),

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transfering mass from the lower to upper size range of the PSD. The increase in volume at sizes larger than 1 μm in Fig. 3a is likely a result of this mechanism; however, uptake of SO_2 as well as the scavenging of unactivated AP by cloud droplets will produce AP with larger masses (Hobbs, 1993)

Precipitation is a cloud processing mechanism that removes AP from the atmosphere (Flossman et al., 1985). Super-micron AP will be the first activated as they are transported from cloud base, i.e. at lower supersaturations, and are the first to grow to precipitable sizes. The near cloud PSD shown in Fig. 3b has no particles larger than 5 μm whereas the cloud base PSD contains particles as large as 20 μm. This is evidence that precipitation has already removed most of the super-micron particles by the time this region of the cloud boundary was measured.

The fourth type of cloud processing pattern is shown in Fig. 3c where the near cloud PSD has virtually the same shape as that at the cloud base, but the concentration at all sizes is smaller by approximately the same fraction. This condition will occur when the ambient air that mixes with the cloudy air has much lower concentrations than cloud base air and the sizes of the cloud base AP have been changed very little by any of the other cloud processing mechanisms.

3. Results

Cloud systems were selected for analysis based on the size of the clouds, their continuity and an inspection of the visual records made with the forward and side-looking video cameras on the aircraft. The acceptance criterion required that cloud lines were isolated from adjoining clouds by no less than 10 km so that far-cloud samples would represent "ambient" AP, i.e. no recently processed particles by clouds. The flights were also classified by AP origin, i.e. continental or maritime. On the days of Flights 7 and 17 the average CN concentrations were significantly higher than the other three flights selected for the analysis (Table 2). This indicates the influence of northerly or easterly winds that bring anthropogenic particles from southern Mexico

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or northern Central America. Flights 9, 12 and 13 were made on days with maritime AP background when winds were from the southwest. Additional confirmation of air mass origin came from back trajectories calculated with the NOAA Hysplit model (http://www.arl.noaa.gov/ready/hysplit4.html). Table 2 summarizes the time of day, location and origin of the AP. The AP origin was determined from the average CN concentration at cloud base. Cases when CN values were less than 500 were considered maritime.

The size of the near cloud region was related to whether these regions were on a growing side of the cloud or the dissipating. Figure 4 shows the relative frequency of the size of near cloud regions. The dissipating regions were on average twice the size of growing regions, with average widths of 4 km and 2 km, respectively.

Each of the near-cloud particle populations were categorized into one or more of the pattern types that were discussed in Sect. 2.4. The 300 m passes were not evaluated for evidence of dilution (Type 4). Multiple classifications are possible, for example the PSD used in the example of Fig. 3a contains both Type 1 and 2 patterns. Forty-eight PSDs were evaluated on clean days and 30 on polluted days. The frequency with which the processing types occurred was evaluated according to altitude, AP origin and whether the near cloud region was on the growing or dissipating side. Table 3 summarizes these results.

Figure 5 illustrates the frequency with respect to the different categories. The region near the growing side of clouds had approximately twice the frequency of dilution processing than the dissipating side (Fig. 5a) whereas the removal of large particle was somewhat more frequent on the dissipating side. The enhanced concentrations of small and large particles (Types 1 and 2) and dilution (Type 4) was most frequently found at 1000 m and 4200 m (Fig. 5b). On the other hand, large particle removal was seldom found at 300 and 1000 m, but was found in 40% of the cases at both 2500 m and 4200 m.

There were very small differences in the frequency of processing types when stratified by AP origin (Fig. 5c). The enhanced concentration of small and large particles

was somewhat more likely for those cases involving continental particles. Large particle removal was more frequent in maritime air masses. In summary, of the 78 cases analyzed, regardless of altitude, AP origin, growth or dissipation, 71% showed enhancement in volume of super-micron particles, 49% had enhanced volumes of sub-micron particles, 24% had super-micron particles removed by precipitation and 28% of the size distributions were classified as homogeneous mixing with boundary layer air.

4. Discussion

The 78 near cloud PSDs were classified unambiguously with one or more of the cloud processing types. The dissipating side of clouds was generally two or more times wider than the growing side but the frequency of cloud processing type appears to have little sensitivity to this characteristic of the near cloud region.

The observations are in general accordance with results from previous observational (Hegg and Hobbs, 1981, 1982) and theoretical studies showing that cloud processing generally shifts the size distribution of boundary layer AP to larger sizes (Flossmann et al., 1985; Flossmann, 1998) by the combined processes of SO_2 uptake and sulfate particle scavenging. Over 70% of the near cloud PSDs showed enhanced concentrations of super-micron sizes and almost 50% had experienced enhancement of sub-micron sizes. The far cloud environments were always lower in AP concentrations than the cloud base AP, suggesting that the enhancement in mass is only a result of aqueous phase chemistry, coalescence, or scavenging, and not interactions with a layer of AP that was advected from a different source than the cloud base AP.

A further evaluation of the frequency of sub and super-micron volume enhancement showed that the enhancement in sub-micron particle volume almost never occurred without enhancement in the super-micron particle volume. This is in qualitative agreement with modeling studies (Flossman et al., 1987; Alfonso and Raga, 2002) that showed that the scavenging of SO_2 by small water droplets and subsequent conversion to sulfate increases the mass of the AP cloud nuclei. In addition, larger water

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droplets have more sulfate in them as a result of the scavenging of SO_2 and the coalescence by smaller droplets into larger ones. The results of Flossmann et al. (1987) also showed that the size distribution of the aerosol particles that remained after the cloud evaporated had shifted to larger sizes than found in the cloud base PSD. This theoretical result is supported by the observational results from this study, as shown in the example of Type 1 and 2 particles (Fig. 3a).

The composition of the AP was not measured in the EPIC observations, but the enhancement in mass can be estimated from the measured size distributions by converting the volume concentrations to mass distributions and assuming that the particles are ammonium sulfate.

Figures 5a–d show frequency diagrams of the difference in particle mass concentration between the near cloud regions and the 300 m environment for sub and super-micron particles, stratified by particle origin. The mass concentration is expressed as a mixing ratio in order to remove the effect of changes in air density with altitude and in order to compare with the modeling results. As observed in Figs. 5a and b, the days with air of marine origin generally had more mass added to the super-micron particles than those of continental origin. The opposite is the case for sub-micron particles that show a slightly higher frequency of mass increase for continental particles. The average increase in mass for sub-micron particles is 0.13 $\mu g\,kg^{-1}$ for particles with continental origin and 0.05 $\mu g\,kg^{-1}$ for the marine case. The average increases in mass for super-micron particles is 1.8 $\mu g\,kg^{-1}$ and 4 $\mu g\,kg^{-1}$ for the continental and marine cases, respectively.

The total, estimated enhancement in sulfate mass of 2 to 4 $\mu g\,kg^{-1}$ is consistent with previous observations and modeling predictions. Measurements of sulfate upwind and downwind of wave clouds over the state of Washington showed enhancements of up to 10 $\mu g\,kg^{-1}$ (Hegg and Hobbs, 1981). Estimates were made of enhancements from sulfate measurements of cloud droplets over the Los Angeles Basin and Western Washington State. Sulfate in excess of ambient values ranged from 0.15–29 $\mu g\,kg^{-1}$. The largest excesses were found in the stratiform clouds over Western Washington State.

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It was speculated that the long lifetimes of these clouds permitted significant uptake of SO_2 that was subsequently converted to sulfate. The Flossmann et al. model (1987) predicted that sulfate in cloud drops could increase from 0 to 4 $\mu g\,kg^{-1}$, depending upon the assumed scavenging rate of AP by cloud droplets, conversion rates of SO_2 to sulfate in the droplets, and the amount of time in cloud. The factor of two differences in mass enhancement of sub-micron particles for the continental and marine cases suggests that the air masses of continental origin probably have higher concentrations of SO_2 that lead to more uptake for these AP in clouds. The factor of two differences in the enhancement of super-micron particle mass indicates the possibility that more coalescence is happening in the marine clouds. Higher concentrations of continental AP lead to higher concentrations of cloud droplets with smaller average diameters and lower probability of coalescence (Twomy, 1974).

5. Summary and conclusions

In situ measurements of aerosol particles in regions around convective clouds within the Mexican ITCZ allow the identification of four types of cloud processing. The most frequent processing mechanism was mass enhancement of sub and super-micron particles, presumably by SO_2 and sulfate particle scavenging. The average increase in mass, estimated from comparisons of far and near cloud size distributions, is consistent with theoretical predictions for marine convective clouds. The comparison of mass enhancements of AP from continental and maritime origins suggest that coalescence in maritime clouds dominates the total enhancement of mass in cloud processed AP.

Cloud processed particles, with increased mass after evaporation, will interact with the environment differently than they would have prior to their transformation. Redistribution and growth to larger sizes will lead to particles that activate at lower supersaturations and grow more quickly to precipitable sizes (Flossmann, 1998). Larger particles scatter light more efficiently and will increase the optical extinction in near-cloud regions. Both of these effects have important climatological ramifications and are the

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focus of ongoing studies.

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Table 1. AP and Cloud Particle Instrumentation on the C130 Aircraft.

Instrument type	Parameter	Range	Accuracy
CN counter TSI Model 3760	Number concentration of aerosol	0.01 to >3 μm to $2\times104\,cm^{-3}$	Varies with concentration, about 6% at $3300\,cm^{-3}$
PCASP	Size spectra of aerosols	0.12 to 3.0 μm 30 channels[1]	±20% Diameter ±16% Concentration
FSSP-300	Size spectra of aerosols	0.3 to 20 μm 30 channels[1]	±20% Diameter ±16% Concentration
FSSP-100	Size spectra of aerosols and cloud droplets	2 to 47 μm 40 channels[1]	±20% Diameter ±16% Concentration
CCN Counter	Number concentration of CCN	0.2 to 1.0% supersaturation	10% at 1% supersaturation
Particle soot/ absorption photometer PSAP	Light-Absorption Coefficient	10^{-7} to $10^{-2}\,m^{-1}$	±5%
Nephelometer M903	Light-scattering Coefficient	1.0×10^{-7} to $10\times10^{-3}\,m^{-1}$	±5%

[1] The PCASP, FSSP-100 and FSSP-300 have had their original electronics replace with the signal processing package (SPP) of Droplet Measurement Technologies that improves response time, eliminates dead-time and increases the number of size channels.

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Table 2. Characteristics of cloud systems selected for analysis.

Flight #	Date 2001	Cloud System	Time Period UTC	Location	Particle Source	300 m CN Conc. cm^{-3}	300 m PCASP Conc. cm^{-3}
7	16 Sept.	1	16:46–17:19	12.3° N, 93.7° W	Continental	910	345
7	16 Sept.	2	18:42–20:12	11.9° N, 95.2° W	Continental	830	227
9	20 Sept.	3	18:16–20:11	10.5° N, 95.9° W	Marine	380	66
9	20 Sept.	4	18:56–20:24	8.2° N, 95.8° W	Marine	200	40
12	28 Sept.	5	17:03–18:12	9.3° N, 93.9° W	Marine	460	138
12	28 Sept.	6	19:14–20:20	11.9° N, 94.1° W	Marine	420	143
13	29 Sept.	7	18:31–19:03	11.4° N, 94.6° W	Marine	360	98
13	29 Sept.	8	19:36–20:22	12.4° N, 94.9° W	Marine	390	64
17	6 Oct.	9	18:34–19:49	11.9° N, 93.9° W	Continental	1900	696
17	6 Oct.	10	20:51–21:36	11.8° N, 94.1° W	Continental	1600	510

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Table 3. Frequency of cloud processed AP types. G = Growing side of cloud, D = Dissipating side of cloud.

Case Type	Altitude m	Number of Cases		Sub-Micron Size Increase %		Super-Micron Size Increase %		Large Particle Removal %		Dilution %	
		G	D	G	D	G	D	G	D	G	D
Continental	300	4	4	0	0	75	0	0	25	0	0
	1000	5	3	80	100	80	100	0	0	20	67
	2500	6	2	50	100	50	100	0	100	33	50
	4200	2	4	100	75	100	100	50	50	100	25
All continental		17	13	53	62	71	69	6	38	29	31
Marine	300	3	9	33	56	67	67	33	0	0	0
	1000	7	5	57	60	86	100	14	0	43	40
	2500	8	4	25	25	38	50	38	75	25	25
	4200	4	8	75	25	100	50	25	50	100	13
All marine		22	26	45	42	68	65	27	27	41	15
Combined	300	7	13	14	38	71	46	14	8	0	0
	1000	12	8	67	75	83	100	8	0	33	50
	2500	14	6	36	50	43	67	21	83	29	33
	4200	6	12	83	42	100	67	33	50	100	17
All altitudes		39	39	49	49	69	72	18	31	36	21
All conditions		78		49		71		24		28	

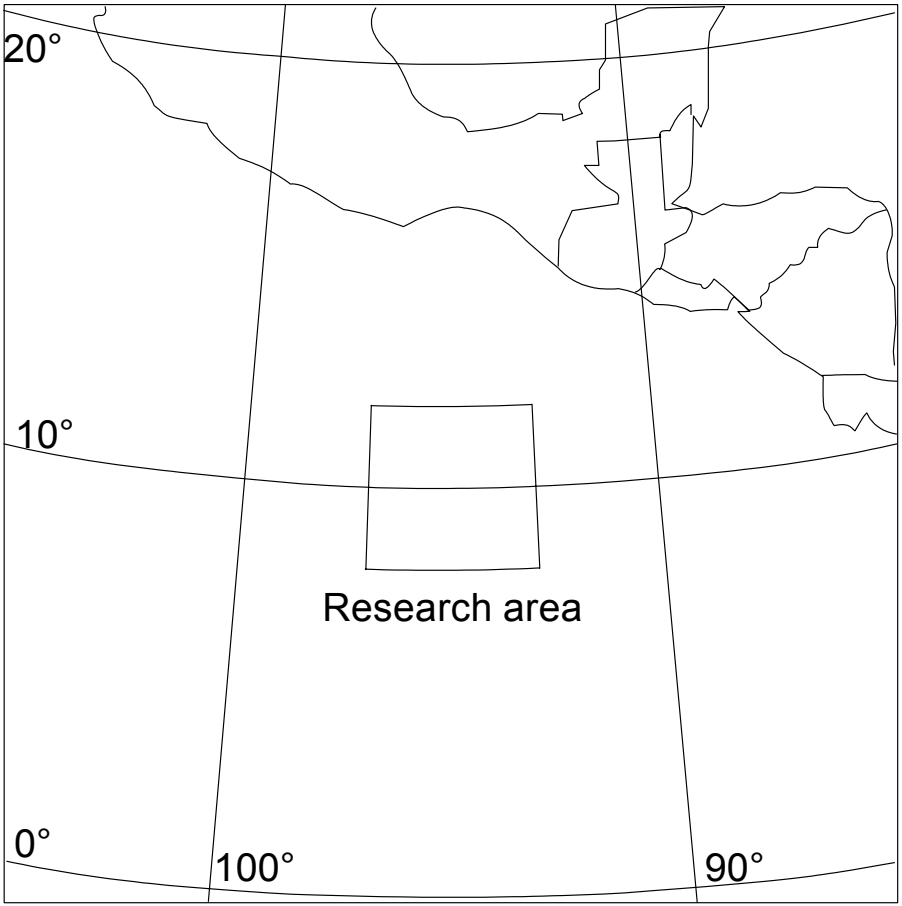

Fig. 1. EPIC-2001 research area during ITCZ flights.

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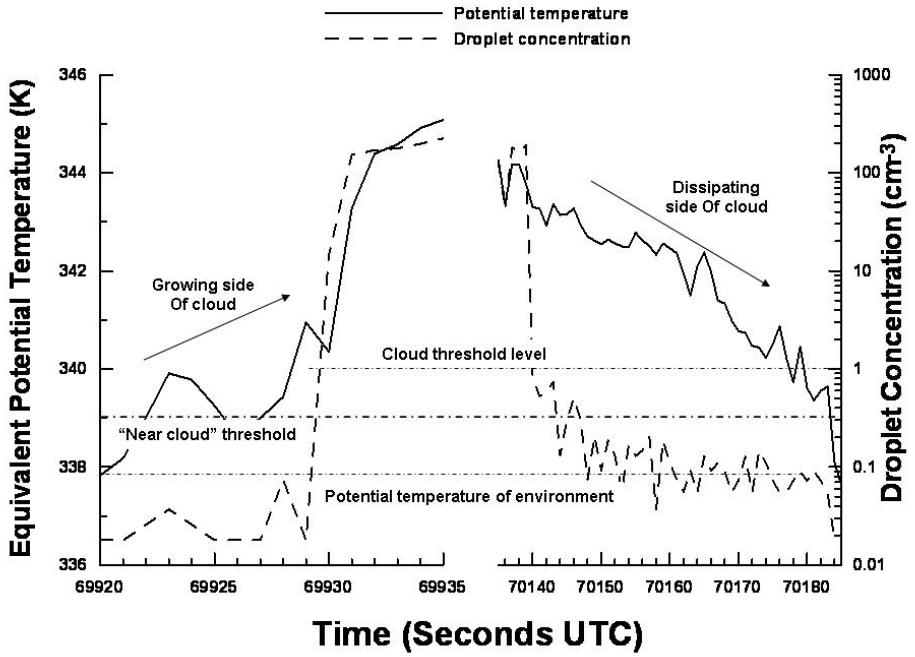

Fig. 2. This time series of equivalent potential temperature solid line and droplet concentration dashed is an example in air near cloud boundaries defined as the FSSP100 droplet concentration >1 cm^{-3}. Air that has mixed with cloud is identified by potential temperature different than the environment 338 K in this example.

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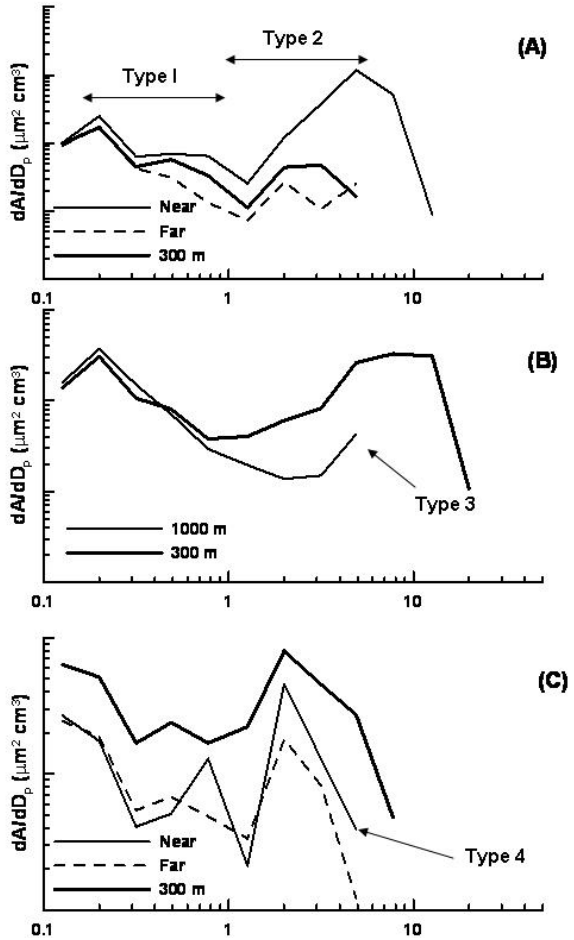

Fig. 3. Size distributions illustrating cloud processing signatures: **(A)** Types 1 and 2, small particle and large particle enhancement, **(B)** Type 3, removal by precipitation, and **(C)** Type 4, dilution with minimal shape change.

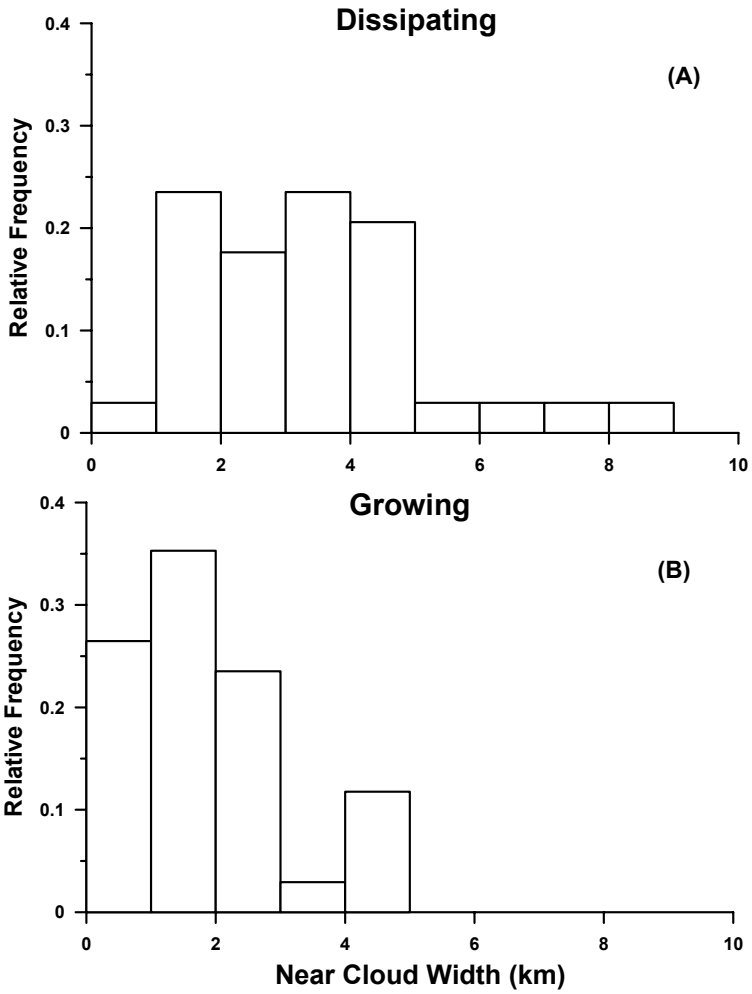

Fig. 4. The relative frequency of the width of near-cloud regions is shown for those sides identified as dissipating **(A)** and growing **(B)**.

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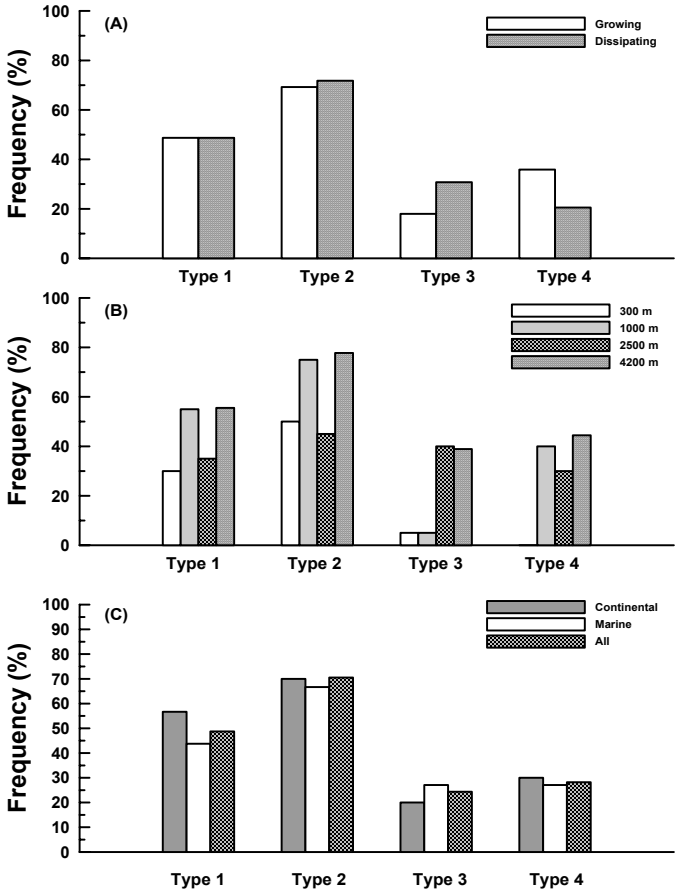

Fig. 5. The frequency of different types of cloud processed AP are summarized here stratified by **(A)** growing or dissipating side of cloud, **(B)** altitude and **(C)** origin of the AP. The 300 m PSDs were not evaluated for type 4 cloud processing.

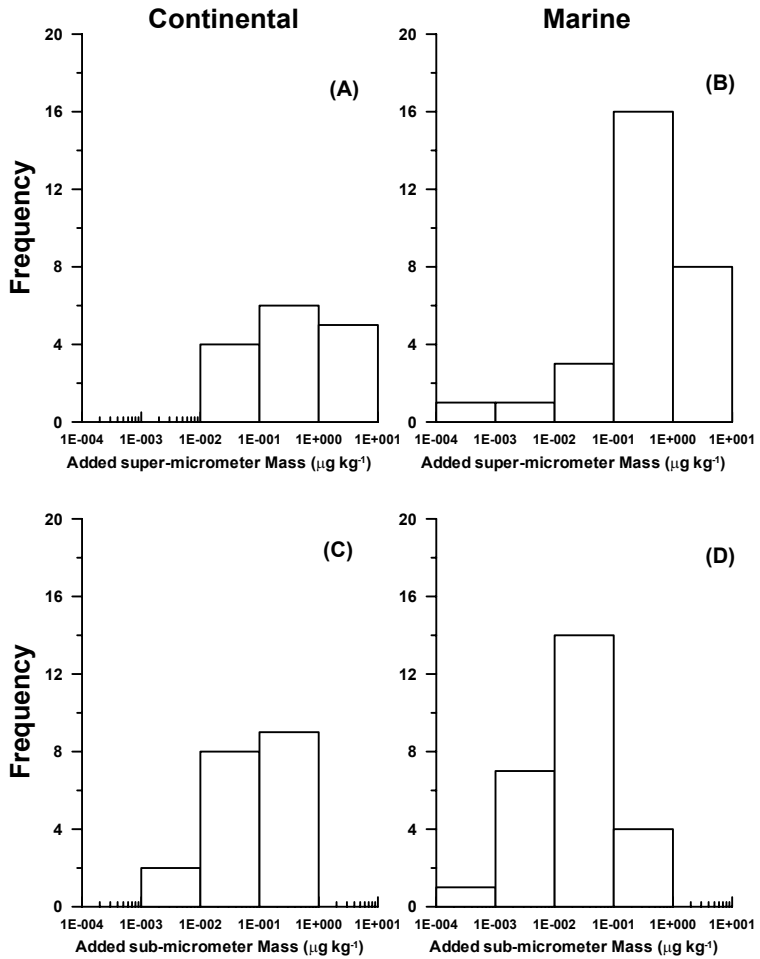

Fig. 6. The frequency of occurrence of differences between near cloud and cloud base AP mass is shown here stratified by air mass origin and particle size range.

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