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Measurement of the Condensation Coefficient of Water in the UMR Simulation Chamber

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Abstract: The UMR Cloud Simulation Facility is described. The facility is designed to provide a controlled environment simulating the conditions of natural atmospheric processes. It consists of two cooled-wall expansion cloud chambers and peripheral instrumentation for generation and characterization of aerosols used for cloud formation studies. Results of initial studies of the growth of warm cloud droplets and inferred measurements of the condensation coefficient are described.

Key words: Cloud chamber, cloud simulation, condensation, droplet growth

Introduction

Over the past 25 years the Graduate Center for Cloud Physics at the University of Missouri-Rolla has been developing a balanced program of experimental and theoretical studies of cloud microphysics. The cloud simulation facility, developed as part of the experimental program, provides a unique opportunity for the study of various processes which occur during the formation and development of clouds under controlled, repeatable, measurable laboratory conditions which simulate natural atmospheric conditions.

Two cooled wall cloud expansion chambers constitute the heart of the facility. A major drawback of traditional expansion cloud chambers has been their short sensitive time due to the influx of heat from the walls following the lowering of the gas temperature by expansion. By externally controlling the temperature of the interior surface of the chamber walls the temperature of the walls and gas can be matched to minimize any heat transfer between the gas and walls as the gas temperature is varied by changes in pressure. In this way a truly isentropic expansion/compression can be carried out at any desired rate from zero up to the maximum design rate of the chamber.

Figure 1 is a block diagram of the air flow through the facility. A continuous source of clean dry air is produced by passing ambient air through a series of filters and driers. The clean dry air is then passed through a precision flowing water humidifier (White *et al.*, 1985) to establish the desired water vapor content before the aerosol to be used for cloud formation is added. The sample is then flushed through one of the two cooled wall expansion cloud chambers (Proto II or Romulus). The aerosol is generated by a four furnace condensation/evaporation method (Alofs *et al.*, 1979). The electrostatic classifier (Knutson and Whitby, 1975a & b) permits extraction of a single narrow size range from the original broad aerosol distribution, and the CFD/HC (continuous flow thermal diffusion/haze chamber) (Alofs, 1978; Alofs and Trueblood, 1981) measures the supersaturation (S_{cr}) at which the aerosol will form a free growing droplet. The alternate gradient CFD (Hoppel *et al.*, 1979) is used to determine the total aerosol concentration in the chamber.

Proto II Chamber

Figure 2 is a line drawing of the smaller Proto II chamber in the 122 cm two section configuration; the chamber is currently operating in the single section, 61 cm high configuration. The chamber is a 10 sided right cylinder with a flat-to-flat inner dimension of 45.7 cm.

The chamber wall consists of a thin (9.5 mm) aluminum inner wall plate and a thick (7.62 cm) outer heat sink with thermoelectric cooling modules (TEM's) between them. The temperature of the interior surface of the inner wall plate is controlled by utilizing the TEM's to pump heat between the inner wall plate and the heat sink as required. Excess heat is removed by circulating fluid from an external temperature controlled circulator bath through passages in the heat sink.

The temperature range of the chamber for a single experiment is from +40°C to approximately 35°C below the heat sink temperature. By lowering the heat sink temperature an overall range of ±40°C can be achieved. The

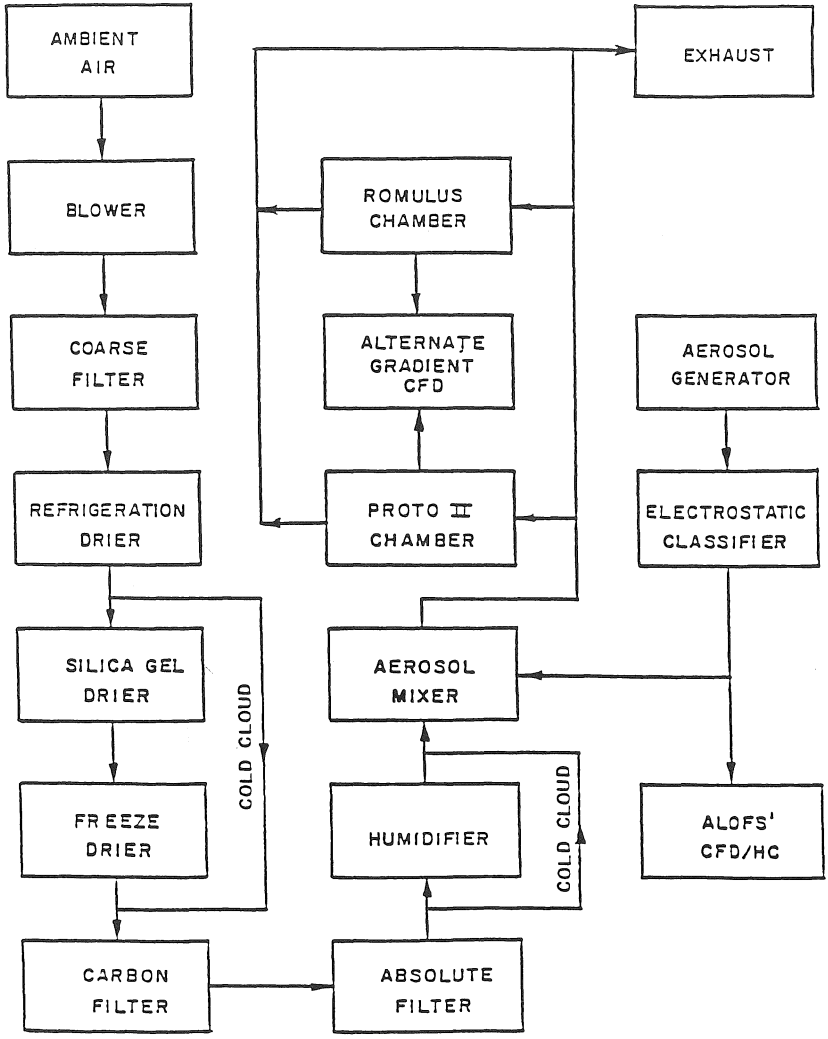

Fig. 1. Block diagram of the air flow through the facility.

maximum cooling rate for the chamber is 10°C/min from +40°C to approximately 10°C below the heat sink temperature and then decreasing as the temperature is further reduced. A maximum heating rate of 10°C/min can be achieved for the entire operating range.

Due to the inherent temperature response lag of the thermally massive walls, the temperature is treated as the independent parameter in designing an experiment. Once the desired temperature *vs.* time profile is selected, a set of

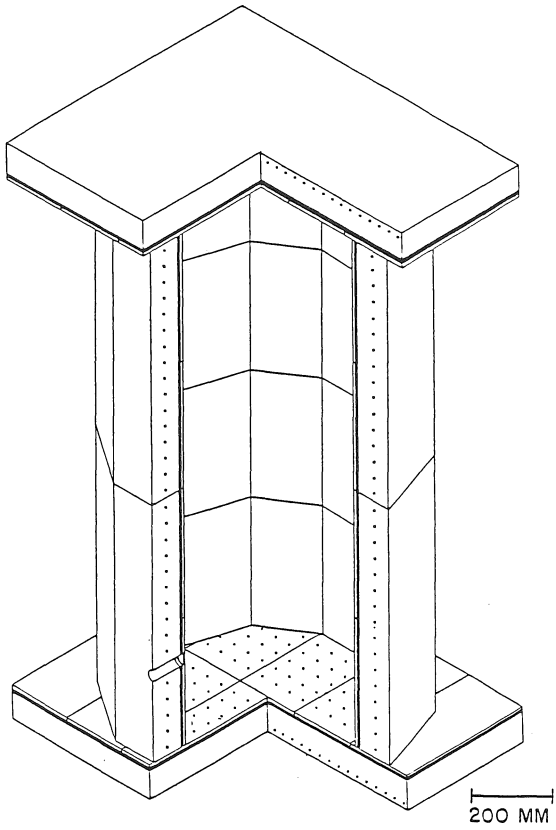

Fig. 2. Proto II Chamber.

aerosol concentrations is also selected covering the range of concentrations of interest. The numerical cloud droplet growth model is then used to calculate the pressure *vs.* time profile required taking into account the latent heat released by the growing drops. This profile is then reproduced by a programmed isentropic expansion/compression cycle in the chamber. A different pressure profile is generated for each aerosol concentration.

During the flushing of the chamber with the sample a small amount is drawn out and the aerosol concentration measured. When the sample in the chamber is determined to be suitable for the planned experiment the appropriate pressure profile is selected for use as part of the chamber closing process. A more detailed description of the wall temperature and chamber pressure control is given by White *et al.*, (1987).

Description of the Experiments

Two experiments are currently being developed. The first is a study of the droplet growth process. Figure 3 is a plot of the temperature *vs.* time and

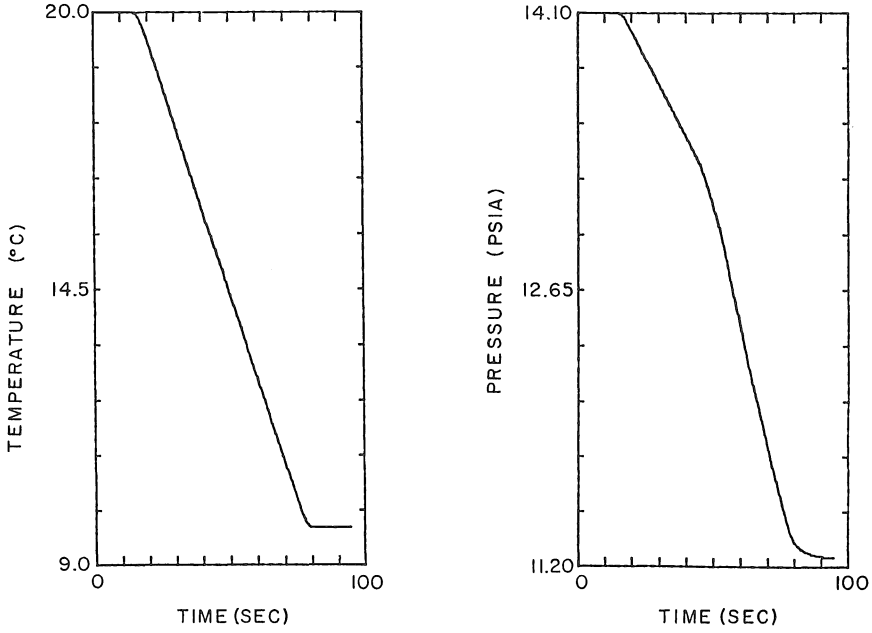

Fig. 3. Control profiles for linear ramp expansion.

pressure *vs.* time control profiles for a simple linear temperature ramp of 10°C/min cooling rate starting at 20°C and cooling to 9.762°C. The knee in the pressure profile, approximately a third of the way through the expansion, is due to the increased expansion rate necessary to compensate for the gas heating due to the release of latent heat caused by cloud condensation. The initial dew point of the sample is approximately 17°C.

In this experiment the development of the cloud droplets as they form and grow on a monodispersed NaCl aerosol is observed and then compared to the numerical cloud growth model. From this data values are deduced for the condensation coefficient, β, (which is the fraction of water molecules striking the droplet surface which stick). A value of this coefficient in the range 0.01-0.1 can account for much of the broadening of the droplet size distribution observed in natural cumulus clouds — a broadening which at present does not have an adequate explanation.

The second experiment is intended to study the effects of droplet growth and evaporation on the scavenging of small noncondensing aerosol particles. This has important application to the mechanisms by which the atmosphere cleans itself.

To extend the growth and evaporation times of a given experiment without creating a cloud of droplets so large that they all fall out onto the floor of the chamber, an oscillating profile such as the one shown in Figure 4 will be used. While the plot shows only one evaporation phase, the main operating program of the control computer is designed to permit a portion of the control profile to

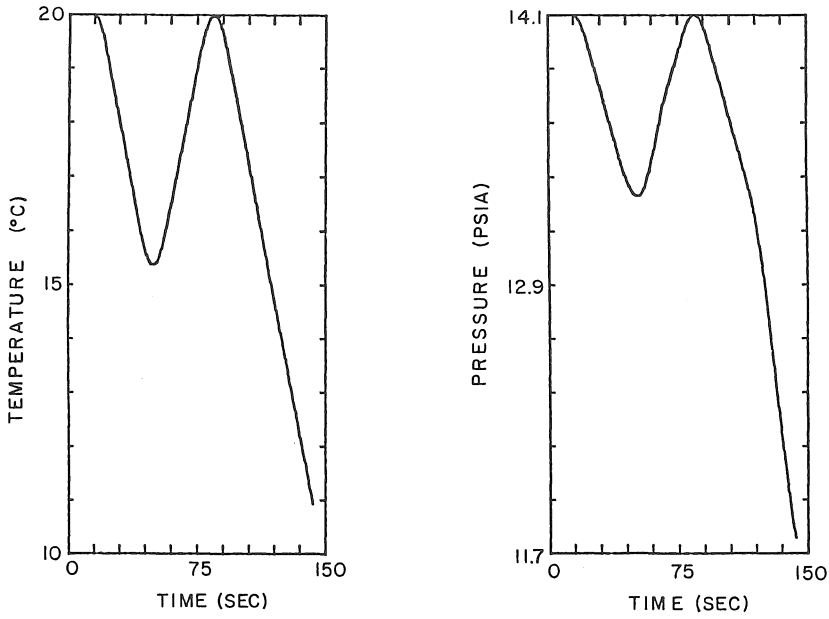

Fig. 4. Control profiles for oscillating expansion.

be repeated a predetermined number of times during a single experiment.

The experimental procedure is based on the use of a sample with two aerosols. One will have an activation supersaturation that will produce a cloud during the bottom part of the oscillations of the control profile; the other, made from a fluorescent laser dye, will not. The first aerosol forms the cloud and the second acts as the scavenged aerosol. After a suitable number of growth and evaporation cycles the cloud droplets will be held at a size of approximately 5 μm while a sample is drawn out of the chamber and examined by a Fluorescence Optical Particle Counter (FOPC). The FOPC detects both the total number of droplets in the sample and the number of droplets which contain a fluorescent dye particle.

Description of the Analysis

Data Collection

Initial expansions for the cloud growth experiment has been performed. At the beginning of each expansion the initial pressure and temperature are recorded, the aerosol concentration is measured and entered together with the measured critical activation supersaturation of the aerosol. During the expansion the computer reads and records the chamber pressure and time at one second intervals.

Droplet size as a function of time is determined by recording the intensity of laser light scattered at approximately 4° to the forward direction. The intensity is

measured by a photomultiplier tube which observes the center of the chamber through a pin-hole aperture which limits its acceptance angle to 1°. A computer generated plot of scattering intensity *vs.* radius of the scattering particle is shown in Figure 5 for light scattered over a range of 1° about a 4° scattering angle.

Local extrema provide a measure of the average radius of the droplets of a monodispersed cloud for discrete values of radius. Overall modulation of the maxima provides assurance that a unique correlation between calculated and measured maxima can be made for each cloud recorded.

During an expansion, the output of the photomultiplier tube is recorded by a multichannel light beam recorder. The recorder also records the shifts of a bistable signal which changes state each time the pressure control is updated. This trace together with the characteristic level shift of the photomultiplier tube trace when a computer controlled shutter is momentarily closed provides an absolute time correlation between the recorder trace and the computer pressure measurements.

The times of the extrema of the scattered light intensity are determined and related to the corresponding droplet radius. This then provides a set of discrete measurements of radius as a function of time. The initial conditions, pressure/time data, and droplet radius/time data constitute input to both of the analysis methods which were used to determine both the initial vapor content of the sample and the condensation coefficient.

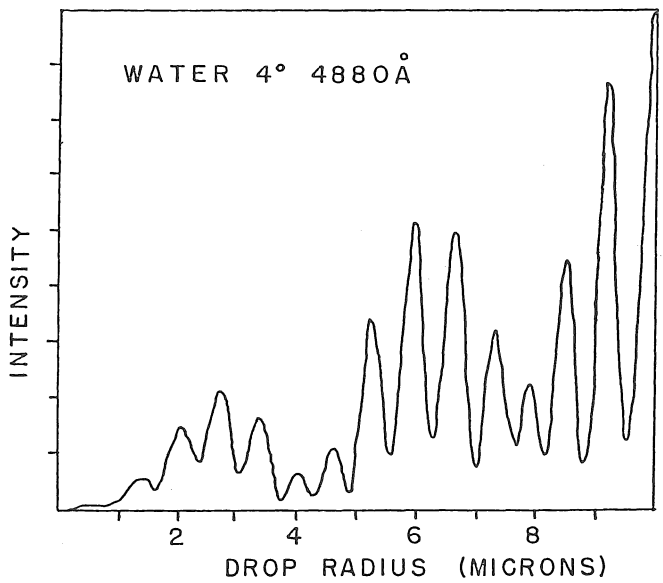

Fig. 5. Computer calculated Mie scattering intensity *vs.* drop radius.

Cloud Arrival Time Method

During a given expansion, the cloud is observable soon after saturation (100% relative humidity) is reached, and this observation provides a good measure of the saturation event. The cloud droplets can be observed starting at 0.70 μm radius. The small time increment (on the order of one second) between saturation and cloud observation, required for drop growth to observable size, can be accounted for in the growth modeling.

The variable chosen to represent water vapor content is mixing ratio, i.e., the number of grams of water vapor per gram of dry air. The essence of the method is to calculate a mixing ratio, r_o' and a condensation coefficient, β, from the observed cloud arrival time and droplet growth rate upon arrival. Let t_1 denote the time at which the supersaturation ratio S reaches unity and t_2 the time at which the drop radius "a" reaches 0.70 μm. The time interval between them is: $\Delta t = t_2 - t_1$.

For expansions with a constant expansion rate, Δt is not sensitive to r_o, but does depend on the condensation coefficient, β. However, its functional dependence, Δt(β), can be extracted from the numerical cloud model (Hagen, 1979). This is accomplished simply by running the cloud model program several times with different values of β, and each time recording the resulting Δt between saturation and a = .70 μm. A polynomial is fitted to these data to get an analytic expression for Δt(β). Given t_2 the expression for t_1 can be written: $t_1 = t_2 - \Delta t(\beta)$.

From t_1 the pressure and temperature can be obtained and an expression for r_o at t_1 deduced knowing that S = 1; then:

$$r_o = \epsilon/[p(t_1)/e_\infty\,(T(t_1)) - 1], \tag{1}$$

where e_∞ denotes the equilibrium vapor pressure over a plane water surface. The standard droplet growth rate formalism (Carstens, 1979) gives an expression for β in terms of the measured initial droplet growth rate a:

$$\beta = (2\pi/R_vT)^{1/2}/[\rho_{sat}(S-S^*)/\rho_w\dot{a} - a/D_{eff}\ell_\alpha LB/KR_vT + (\pi/2R_vT)^{1/2}] \tag{2}$$

where R_v is the gas constant for water vapor, T the temperature, ρ_{sat} the saturation water vapor density over a flat surface, S* the equilibrium supersaturation ratio over a droplet of radius (a), ρ_w the density of liquid water, D_{eff} the effective diffusion coefficient for water vapor in air (Carstens, 1979), ℓ_α the length associated with thermal accommodation (Carstens, 1979), L the latent heat of condensation for water, $B = de_\infty/dT$, and K the thermal conductivity of moist air. Note that r_o is implicitly contained in Equation (2) in S, and that β is implicitly in Equation (1) through t_1. Inserting Equation (2) into Equation (1) yields one equation and one unknown, r_o. Its solution using the experimental data t_2 and a yields r_o.

Droplet Growth Method

The basis of this method is to calculate r_o (and β) from two or more measurements of a. The droplet growth rate equation (Carstens, 1979) for the i-th droplet growth rate measurement can be written

$$r_o = \epsilon/[p_ie_\infty(T_i)^{-1}/\{(a_i+\ell_i)\dot{a}_i/D_{eff}\,\rho_{sat,i} + S_i^*\}^{-1}] + 4/3)\pi Na_i^3, \tag{3}$$

with $\epsilon = .62197$, p is the total pressure, N is the droplet concentration, and

$$\ell = D_{eff} (\ell_\alpha \, LB/K \, R_v \, T + \ell_\rho/D)$$

$$\ell_\beta = (1/\beta - 1/2) \, D \, (2\pi/R_v T)^{1/2}.$$

Here D is the diffusion coefficient for water vapor in air. The amount of water in the system is constant, so Equation (3) should have the same value for all drop growth measurement points. Define a minimization parameter as:

$$\text{chisq} = \frac{\sum_{i=2}^{I} \sum_{j=1}^{i-1} (r_{oi} - r_{oj})^2 / [\delta(r_{oi} - r_{oj})]^2}{\sum_{ij} [\delta(r_{oi} - r_{oj})]^{-2}},$$

where $\delta(r_{oi} - r_{oj})$ denotes the uncertainty in the knowledge of $r_{oi} - r_{oj}$. This uncertainty is calculated based on a .05 μm uncertainty in "a". a 20% uncertainty in $\dot{a}$, and a 5% uncertainty in N. The condensation coefficient enters into chisq through ℓ. Since r_o is constant, chisq should be zero or at least small. Using a standard computer optimization routine, β can be varied to minimize chisq, yielding a value for β which can be put back into Equation (3) to get r_o.

Recall that the cloud arrival time method required a constant expansion rate, to make Δt independent of r_o. For the multiple growth rate method there is no such requirement.

Results

Figures 6, 7, 8 and 9 are plots of the measured condensation coefficient *vs.* droplet radius (time) for data taken on four separate days. Data from each individual expansion on a given day is shown by a different type of mark on the plot.

An examination of the data shows that β decreases as the experiment proceeds, that is as time progresses or the droplet radius increases. The spread in the values of β amongst several identical expansions on the same day corresponds to an uncertainty of 0.00002 in the water vapor initial mixing ratio. During the latter stages of the cloud development the value of β drops to a sufficiently small value to influence droplet growth. This could cause broadening of the droplet size distribution, and is probably responsible for the observed distribution at cloud bases in the atmosphere (Manton and Warner, 1982).

Data from a series of 5°C/min expansions gave similar results of β as a function of droplet size. Passing the sample air through an activated charcoal filter does not produce any observable change.

A set of preliminary multiple expansion profiles were carried out and the initial analysis of the data shows that the value of β for the second formation of the cloud starts at a value equal to or less than the smallest value observed during the first formation of the cloud prior to evaporation. During the second growth the measured value of β continues to decrease to a final value between 0.01 and 0.02.

At this point the changing value of β is treated as an observed phenomenon and is presented without attempt to explain it.

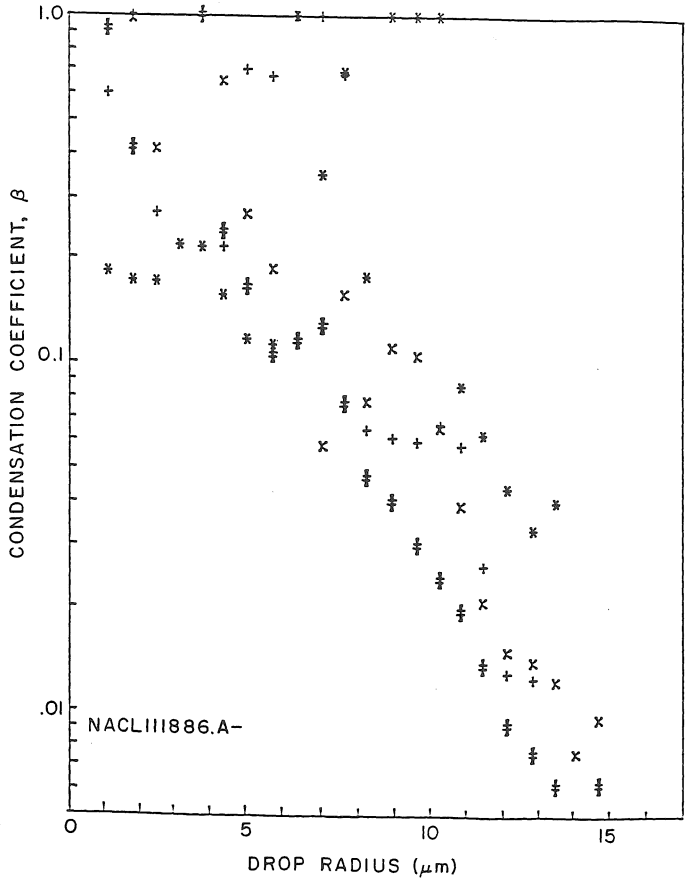

Fig. 6. Data from 18 November 1986.

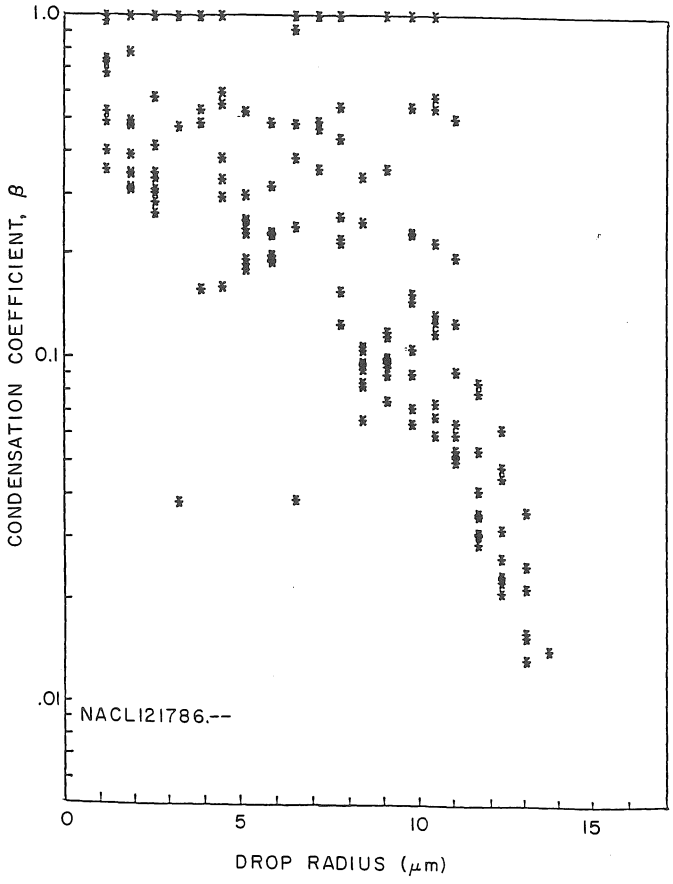

Fig. 7. Data from 17 December 1986.

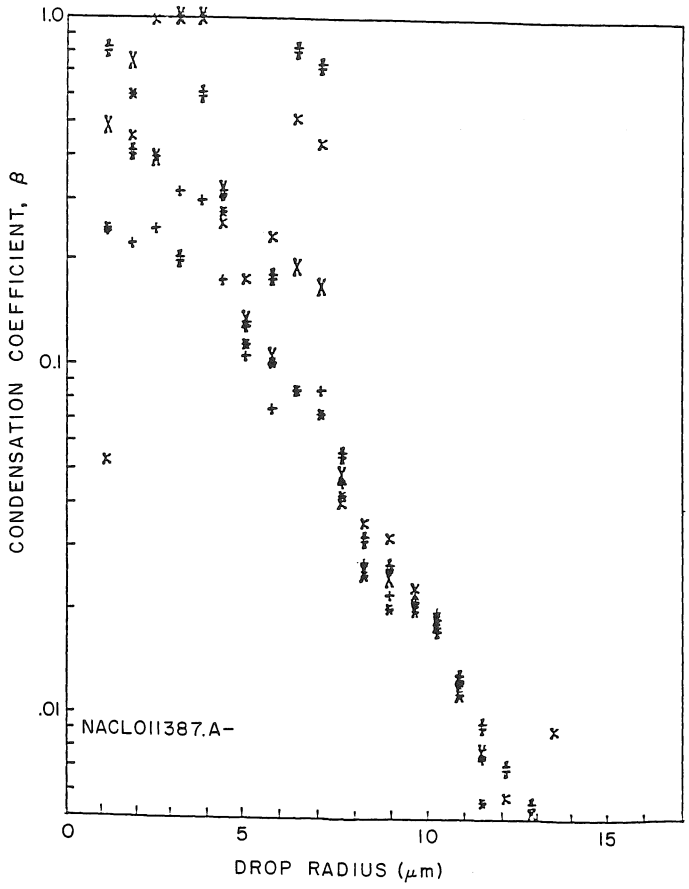

Fig. 8. Data from 13 January 1987.

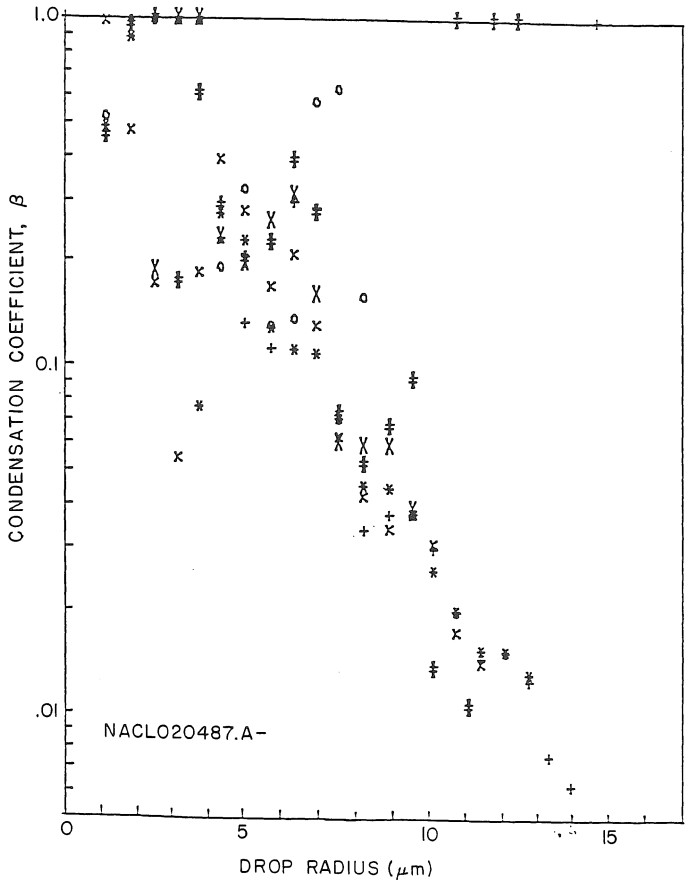

Fig. 9. Data from 4 February 1987.

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