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MEASUREMENT OF THE CONDENSATION COEFFICIENT OF WATER IN  
THE UMR CLOUD SIMULATION CHAMBER

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ABSTRACT

Measurements of the condensation coefficient of water under conditions closely approximating those in natural atmospheric cloud have been made in the cooled-wall UMR cloud simulation chamber. Current measurements disclose a value of condensation coefficient near unity at the outset of the experiment, generally decreasing to lower values ( $\sim .01$ ) as the experiment progresses. The significance of the magnitude of condensation coefficient in atmospheric cloud is briefly discussed.

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I. INTRODUCTION

The measurement of the condensation coefficient of water is important in a number of areas. We approach this problem from the cloud physics side, so our concern is with the dropwise condensation of water under normal atmospheric conditions. We begin by a few definitions and a brief discussion of why we regard the condensation coefficient to be significant in Cloud Physics.

The condensation coefficient,  $\beta$ , may be defined as the probability that a water molecule, upon striking a liquid water surface, sticks to that surface.

Condensation growth of a cloud drop is accompanied by heat conduction (due to the latent heat of condensation), and the thermal accommodation coefficient,  $\alpha$ , is the thermal counterpart to  $\beta$ . It represents the extent to which "air molecules" accommodate to the temperature of the liquid surface (or the drop)

$$\alpha = \frac{E_i - E_r}{E_i - E_S}$$

where  $E_i$  = ave. energy/molecule of incoming stream  
 $E_r$  = ave. energy/molecule of reflected stream  
 $E_S^*$  = ave. energy/molecule of a stream corresponding  
to surface temperature.

The most familiar form of drop growth theory is that adapted to the low supersaturations found in atmospheric clouds (e.g. Fitzgerald, 1970; Fukuta and

Walter, 1970; Carstens, 1979). Carstens' version may be written to reasonable approximation as follows:

$$(a + \lambda) \frac{da}{dt} = D_{\text{eff}} \frac{\rho_{\text{eq}}(T_{\infty})}{\rho_{\lambda}} [S(t) - S_{\text{sat}}(a)] \quad (1)$$

where

$$\lambda = D_{\text{eff}} \left( \frac{\lambda_{\beta}}{D} + \frac{\lambda_{\alpha} \tilde{\text{Ln}}_g}{pK} B \right), \quad (2)$$

$$\frac{1}{D_{\text{eff}}} = \frac{1}{D} + \frac{\tilde{\text{Ln}}_g}{pK} B, \quad (3)$$

$$\lambda_{\beta} = \frac{4D}{\bar{v}} \frac{1}{\beta}, \quad (4)$$

$$\lambda_{\alpha} = \frac{8K}{n_g R \bar{v}_g} \frac{\gamma-1}{\gamma+1} \frac{1}{\alpha}, \quad (5)$$

and where  $S_{\text{sat}}(a)$  is normally written

$$S_{\text{sat}}(a) = 1 + \frac{r^*}{a} - \frac{Am_{\beta}}{a^3} \quad (6)$$

where  $r^*$  and  $A$  are constants, and  $m_{\beta}$  is the mass of dissolved salt (e.g. Rogers, 1976). Here  $\rho_{\text{eq}}(T_{\infty})$  represents the ambient saturation vapor density;  $B$  is the slope of the vapor saturation curve at ambient temperature (i.e.  $B = de_g/dT_{\infty}$ ); and the remaining terms are defined in Appendix A.

## II. IMPORTANCE IN DROP GROWTH

### A. Influence on growth rate

An important question is the following: "How rate influencing is the accommodation process, represented by the above parameters, in drop growth?" Under ordinary atmospheric conditions where water vapor is a dilute species with respect to air, one normally assumes that diffusion and conduction are the major rate controlling

steps in condensation growth. Plainly, however, if  $\beta \rightarrow 0$  the condensation probability takes over as the rate controlling step. Condensation rates are rather different depending on which process predominates. The matter may be put briefly by looking at the "resistance" term in the transport of vapor molecules to the surface of a drop of radius  $a$ .

Suppose for the sake of argument we ignore latent heat in the formalism represented by equation(1). Then  $D_{\text{eff}} = D$ , and (1) can be rewritten

$$a \frac{da}{dt} = \frac{\rho_{\text{eq}}(T) [S(t) - S_{\text{sat}}(a)]}{\rho_l \frac{1+\lambda/a}{D}}$$

where  $(1+\lambda/a)/D$  can be regarded as a resistance. For diffusion controlled growth this resistance is given simply by  $\frac{1}{D}$ ; for growth controlled jointly by diffusion and surface accommodation this resistance may be written approximately by:

$$\frac{1 + \frac{\lambda}{a\beta}}{D} = \frac{1 + \frac{\text{Knudsen number}}{\beta}}{D}$$

where  $\lambda$  is the mean free path of diffusant (vapor) molecule. Thus if either  $\beta \rightarrow 0$  or  $a \rightarrow 0$  the process tends to be dominated by surface kinetics.

#### B. Significance in Cloud Physics

One of the nagging problems in cloud physics has been the repeated observation that cloud drop size distributions are broader than conventional (closed parcel) theory implies. The most popular explanation appeals to mixing, but inexplicably broad size distributions have also been observed (Jensen and Baker, 1986) for unmixed, i.e., adiabatic, parcels. In the unmixed case the value of  $\beta$  is of significance, for if it is sufficiently small it is capable of accounting for that portion of the distribution extending to the small-size end of the spectrum (Warner, 1969).

### III. THE SEARCH FOR $\beta$

#### A. Existing measurements of $\beta$

The problem with which we are here concerned arises in establishing an experimental value for  $\beta$ . Past measurements disclose a scattershot of values ranging from 0.001 to 1.0, with cloud physicists often taking a value of around 0.036 as a sort of compromise (Pruppacher and Klett, 1978). It is in fact hard to justify a choice for  $\beta$  based on existing measurements. The general consensus among researchers is that the condensation coefficient for a "pure" surface ought to be unity (Mozurkewich, 1986); a common hypothesis involved to "explain" the variety of values

actually measured is that trace contaminants may strongly affect such measurements. Thus the existence of trace contaminants in the real atmosphere may mean that the condensation coefficient is significantly low under ordinary meteorological conditions.

#### B. The UMR program

There would seem to be little point in simply adding another value of  $\beta$  to the variety already in the literature. Yet this variety attests to the importance of the phenomenon and entreats a resolution to the problem. We have therefore started a systematic program aimed at measuring the condensation coefficient of water over a broad range of carefully controlled conditions. We have adopted the hypothesis that departures of  $\beta$  from unity ( $\beta < 1$ ) are attributable to contamination. Hence an important part of our program will consist of precautions to ensure system purity, as well as subsequent deliberate introduction of selected "contaminants". In addition ambient supersaturations will be varied over a broad range, starting with magnitudes typical of atmospheric cloud, ( $\sim .003$ ) and extending to values approaching those associated with homogeneous nucleation ( $S \sim 4.0$ ). High supersaturations are produced in the UMR fast expansion chamber. Low values are produced in the UMR cloud simulation chamber. The program has been started with the simulation chamber.

### IV. MEASUREMENT OF $\beta$ IN THE UMR SIMULATION CHAMBER

#### A. OVERVIEW OF THE FACILITY

The simulation facility consists of a dynamic cooled-wall expansion chamber which simulates in-cloud thermodynamics. The hallmark of this device is the close instantaneous tracking of its wall temperature with respect to that of the interior gas. This dynamic tracking suppresses the free convection that would otherwise occur at the walls, and permits prolonged experiments under well controlled conditions.

Sample preparation consists of (1) careful filtering of outside air, followed by (2) humidification, and (3) introduction of a prepared aerosol consisting of cloud condensation nuclei. This sample is introduced into the chamber where it is subjected to an isentropic expansion. The resulting cloud is analyzed by direct photography, laser scattering, or sample withdrawal.

#### B. Sample preparation

Preparation begins by drawing in outside air (1-2 l/sec) and passing it through a coarse surgical cotton filter. Water vapor is removed by passing the air through a

refrigerant dryer ( $\sim 0^{\circ}\text{C}$ ), followed by a silica gel dessicant column and low temperature ( $- 40^{\circ}\text{C}$ ) freeze dryer. Finally the air passes through a column of activated carbon granules (to remove residual organics), followed by an absolute filter rated at 99.97% efficient at  $0.3 \mu\text{m}$ .

The clean dry air is then humidified by passing it through a pair of humidification columns. (The dew point of the air at  $17^{\circ}\text{C}$  is consistently  $0.355^{\circ}\text{C}$  below the outlet temperature.)

Finally a prepared aerosol, monodisperse NaCl nuclei in these experiments, is introduced into the clean moist air sample at a mixing ratio of 1:100 or less. At this point the sample is ready to be introduced into the simulation chamber.

#### C. Sample introduction

The sample is introduced into the chamber from top to bottom. It first enters the inlet manifold chamber above the top and then is introduced uniformly into the chamber by means of 164 distinct ports located between the thermoelectric modules (so as not to interfere with their operation). This inlet system is duplicated as an outlet system at chamber bottom.

The chamber is flushed for a minimum of 15 minutes and continues until a measurement of the aerosol concentration shows an acceptably stable value for 5 min. When all systems are operating normally the chamber is sealed by closing the inlet and outlet valves. After a period of equilibration the chamber is ready for a cloud-generating expansion.

#### D. Aerosol preparation

Part of the clean dry air sample is used as a carrier gas for the four furnace NaCl aerosol generator (the procedure is described in some detail by Alofs et al., 1979). A boat containing granulated NaCl is placed in the first furnace region ( $\sim 730^{\circ}\text{C}$ ). This gives off NaCl vapor which, when cooled ( $\sim 300^{\circ}\text{C}$ ) in the region between furnaces # 1 and 2, forms an aerosol by homogeneous nucleation. Subsequent reevaporation and condensation narrows the size distribution (Kitani and Ouchi, 1967), producing mean sizes with diameters between 0.01 and 0.08 microns. (Geometric standard deviation  $\sim .12$ ; CCN concentrations approach  $10^5/\text{cm}^3$ .)

The NaCl aerosol is further narrowed by passing it through a TSI classifier. Once a monodisperse CCN is produced, it is immediately introduced into the clean moist air system prior to entering the simulation chamber. Chamber concentrations currently

range from 25 to 150 CCN/cm<sup>3</sup>.

#### E. Isentropic cooling

The desired time-temperature-pressure control profile is loaded into the control computer. The computer uses this profile to generate a signal to the wall controllers and the expansion valve of the chamber.

#### Wallcooling

Wall temperature control is based on the use of thermoelectric modules to pump heat between the thin (0.96 cm) inner wall and thick (7.6 cm) fluid-thermostated outer wall. Wall temperature is controlled by separate analog controller circuits for each 15.25 x 30.5 cm control section of wall. Each controller receives two inputs, one from the output of a thermometer mounted on the wall section and the other from a control computer. Based on a comparison of these signals, the controller outputs a command signal to the programmable switching power supply which then causes the wall section to heat or cool so as to drive the difference signal to zero.

#### Expansion

The expansion consists of the controlled removal of air radially from the sensitive volume through a series of small ports (16 ports, 0.79 mm dia. per 61 cm height) located between side wall heat sink sections. These ports are collected into a channel which is connected to an expansion manifold. An eight bit digital valve controls flow into or out of the chamber. This valve is connected to a vacuum ballast tank (for expansion) which in turn is connected to a continuously running 45 l/min. mechanical vacuum pump.

Pressure is the measured variable during the isentropic expansion. It is measured by a differential strain gauge pressure transducer with its reference side connected to the sensing volume of a vacuum-referenced dead weight gauge.

The TEM's can cool the inner wall surface at rates up to 10°C/min for temperatures from + 40 to 10°C below the temperature of the heat sink. Below this range the maximum cooling rate decreases until the lowest temperature of 35°C below the heat sink temperature is reached. Maximum heating rate exceeds 10°C/min for all temperatures. At present the chamber at 20°C regularly shows an rms spread of 0.01°C or less in the temperature of the 40 measured control sections with a peak-to-peak spread of less than 0.050°C. For the interior walls 30°C below the heat sink temperature, the rms increases to 0.075°C and the peak-to-peak spread approaches 0.5°C.

### Observation and sizing: Mie scattering

Because of this wall-tracking feature the walls are necessarily opaque. Access to the chamber is provided by three 2.3 cm diameter windows: two directly opposite each other and one at 72° from the forward direction. Each window is 1.6 mm thick Sapphire coplanar with the inner wall. About 4 mm behind the coplanar window another window (or other element) can be placed. Temperature control is achieved by passing CO<sub>2</sub> gas between plates. A transistor thermometer sensor is in contact with the edge of the window and its signal used for control.

The window at 72° provides a port either for photography or low light level TV camera. Photographic illumination is provided by a Xenon flash lamp. Its beam is shaped with lenses and a slit into a vertical sheet of light in the chamber. All drops in the light are in focus for the camera lens and these are registered on film. TV illumination is provided by the laser beam.

Actual measurements of drop sizes are made by Mie scattering. The 4° forward pattern is sufficiently structured so as to identify specific peaks with particular drop radii. Thus a series of peaks and valleys permit one to directly infer drop radius vs time for a monodispersed cloud.

### Results

The condensation coefficient was inferred directly from drop growth rates using continuum theory. (We have avoided the temptation to lower pressure, so as to enhance the role of surface kinetics, because we preferred not to rely on theories in the transition regime of Knudsen number.) The theory is basically that presented by Carstens (1979); it is similar to the often-quoted theory of Fukuta and Walter (1972), except with convective mass flux (Stefan flow) and enthalpy flux included.

Preliminary data has now been taken. All the data show a rather large spread in the vicinity of  $\beta = 1$  for the early stages of growth, out to about 6 micron radius. Thereafter the value of  $\beta$  drops monotonically reaching values as low as 0.005 near radii of 15  $\mu$ . These data were generated by subjecting a monodisperse aerosol to linear "ramped" cooling rates of 10°C/min. Figure 1 illustrates the trend of this data for several 10°C/min runs.



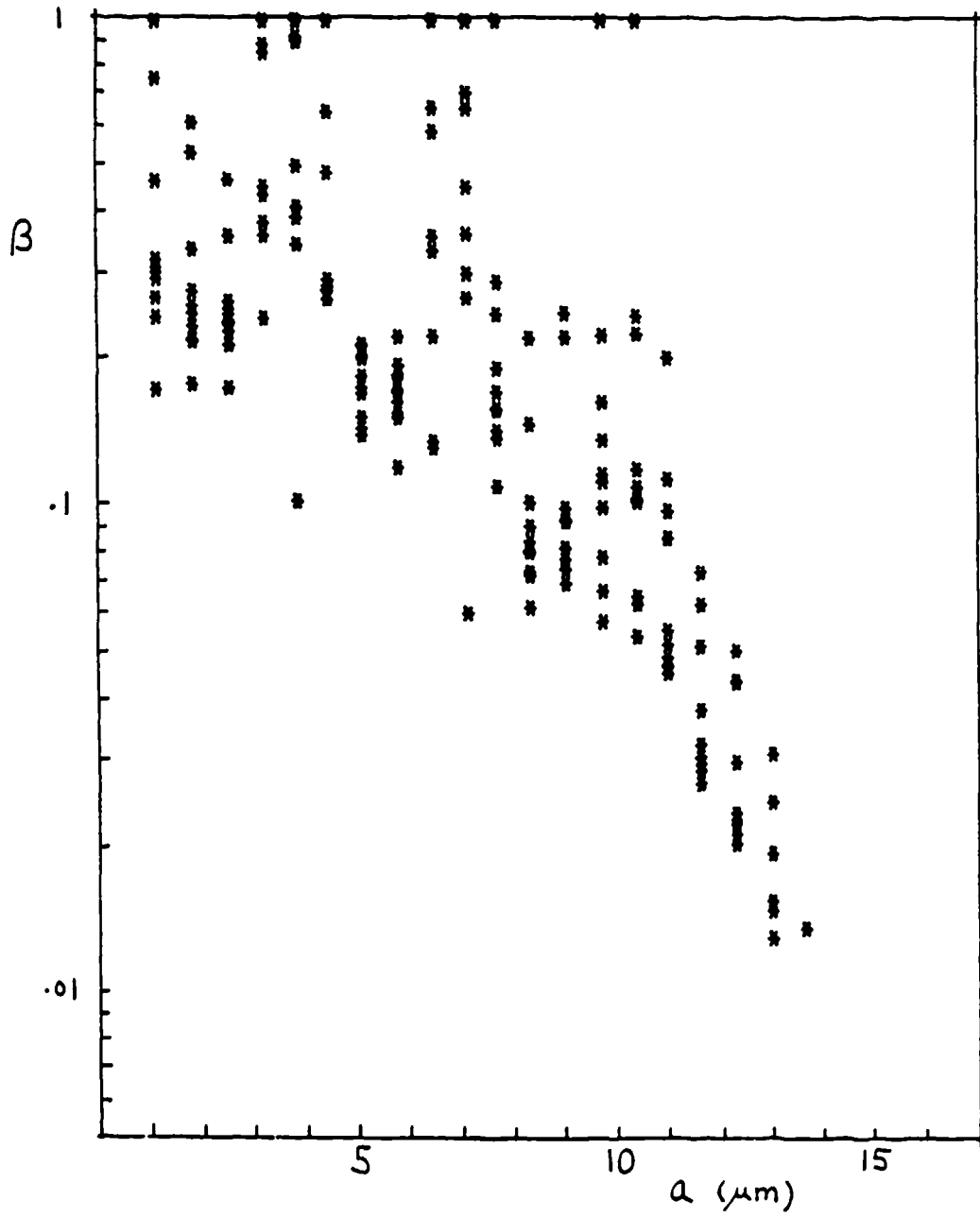


FIGURE 1

## APPENDIX A: LIST OF SYMBOLS

In the following list the tilde over a variable designates it as a molar quantity, e.g.,  $L$  denotes latent heat in cal/gm, and  $\tilde{L}$  cal/mole.

### English symbols

$a$  drop radius  
 $D$  diffusion coefficient, water vapor in air  
 $e_s$  vapor pressure  
 $K$  thermal conductivity, air  
 $L$  latent heat of condensation  
 $n_g$  molar concentration, air (moles/cm<sup>3</sup>)  
 $p$  pressure  
 $R$  universal gas constant  
 $S$  supersaturation ratio  
 $T$  temperature  
 $\bar{v}_g$  average speed, gas molecule

### Greek symbols

$\alpha$  thermal accommodation coefficient  
 $\gamma$  ratio of specific heats (const. pressure: const. vol.)  
 $\rho_l$  density (gms/cm<sup>3</sup>), liquid water

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