

Operation of a Thermal Diffusion Chamber for Measurements on Cloud Condensation Nuclei¹

V. K. SAXENA, J. N. BURFORD AND J. L. KASSNER, JR.

Graduate Center for Cloud Physics Research, University of Missouri, Rolla

(Manuscript received 25 September 1969)

ABSTRACT

In recent years the thermal diffusion chamber has found wide application in studying condensation nuclei (CCN) which are effective in natural cloud and fog formation. Explicit solution of the equations governing the transient behavior of the chamber suggests the necessity for precise control of the temperature or the relative humidity of the incoming sample if meaningful measurements are to be obtained. A recent conclusion of Fitzgerald, that transient supersaturations exceeding the steady-state peak value may arise if the incoming sample is saturated at a temperature less than that of the (hot) top plate, is also verified. Further, it is pointed out that as long as turbulence occurs while introducing the sample, the transient behavior of the chamber remains indeterminate. This defect may be eliminated by giving special consideration to the method of sample introduction. The measurements of CCN concentration as a function of time at Rolla taken with a thermal diffusion chamber are also presented. The CCN concentration, in general, is found to follow local meteorological conditions. The trend of our data shows a qualitative agreement with that available in the literature. It is suggested that an intercomparison of the data taken on a carefully fabricated thermal diffusion chamber with those taken on the other types of cloud chamber would help to decide the potential value of the former as a CCN counter.

1. Introduction

Condensation nuclei are generally classified according to their size, those having radii in the range 10^{-7} – 10^{-5} cm being called Aitken nuclei and those with radii $\geq 10^{-5}$ cm (up to 10^{-4} cm) usually being referred to as cloud condensation nuclei. Indeed, these dimensions lie outside the range of the resolving power of an ordinary microscope and this classification does not seem to have much practical meaning. Another important and more practical characteristic by virtue of which these nuclei can be classified is their effectiveness in activating the condensation of water droplets at different supersaturations. From this point of view those nuclei which activate at supersaturations in the range normally encountered in natural cloud (Warner, 1968) and fog (Kocmond and Jiusto, 1968) formation may be classified as cloud condensation nuclei (referred to hereafter as CCN). The investigation of various properties of CCN requires the production, maintenance and control of low supersaturations. This has been achieved most readily in two types of diffusion cloud chambers (Vollrath, 1936; Langsdorf, 1936, 1939).

The chemical gradient cloud chamber discovered by Vollrath employs the diffusion of two chemically different vapors at a constant temperature. The thermal gradient or thermal diffusion cloud chamber of Langsdorf produces a supersaturation by non-isothermal dif-

fusion of vapor under the influence of a temperature gradient. The former type of cloud chamber has been successfully employed (Twomey, 1959a,b, 1960) for the study of CCN. However, the thermal diffusion chamber affords an unusual and unique opportunity of producing a wide range of supersaturations depending upon the thermal gradient employed and is thus well suited for the study of both homogeneous (Franck and Hertz, 1956; Katz and Ostermier, 1967) and heterogeneous (Schiff *et al.*, 1953; Wieland, 1956) nucleation. On account of its ease in fabrication and simplicity of operation, it has found wide application in investigating the supersaturation spectrum of CCN (Twomey, 1963), which is related to the size distribution in a complex and not very well understood way (Twomey, 1965). It has also been used as a CCN counter (Squires and Twomey, 1966; Twomey and Warner, 1967; Jiusto, 1966; Jiusto and Kocmond, 1968a; Radke and Hobbs, 1969a,b; Ohta and Uchida, 1969; Twomey and Wojciechowski, 1969), as well as for measurements of the activation supersaturation of nonhygroscopic nuclei (Jiusto and Kocmond, 1968b). This latter usage provides a partial test of the Volmer (1939) theory of nucleation on insoluble particles. The effectiveness of fire as a source of CCN has been studied (Hobbs and Radke, 1969) using the thermal diffusion chamber as a CCN counter. The chamber has also been used (Twomey, 1968) for studying the volatility of the CCN. Apart from CCN, there exists in the atmosphere some large and giant cloud nuclei which activate even at

¹ Research supported by the Office of Naval Research under THEMIS Contract No. N00014-68-A-0497.

slight subsaturations and thereby play an important role in fog formation (Kocmond and Jiusto, 1968; Kocmond and Pilie, 1969). Such subsaturations have been produced by Kocmond and Jiusto in a haze chamber which is essentially a modified thermal diffusion chamber.

Accompanying the increasing popularity of the thermal diffusion chamber as an important tool in cloud physics, some improvements (Gagin and Terliuc, 1968) and modifications have been suggested in its design and operation from time to time. Storozhilova (1961) was the first to suggest a continuous flow thermal diffusion chamber which ingeniously operates on a dilution technique of the test sample, rendering it a reliable CCN counter at large supersaturations and high concentrations. In addition, the effect of vapor depletion is reduced in this type of chamber. Severynse (1964) employed two vertical concentric cylinders instead of two horizontal plates, with a horizontal temperature gradient and thus allowed the natural convection currents in the chamber to keep the droplets in a region of high supersaturation for an extended period of time to enhance droplet growth. This chamber has been stated by Severynse to approximate more closely the conditions encountered in natural cloud formation, and was indeed employed by Twomey and Severynse (1964) for investigating the size distributions and supersaturation spectra of natural aerosols. Laktinov (1968) further improved the nuclei counting device and used this chamber for studying the aerosols contained in the surface boundary layer (Laktinov, 1967). He arrived at an interesting conclusion that in the types of aerosols investigated there exists no clear correspondence between the distribution of aerosol particles by size and the distribution of cloud nuclei by supersaturation. In fact, this is what renders the classification of condensation nuclei with respect to their activation supersaturation more practicable. Even the photographic counting method generally used in these cloud chambers has been questioned (Laktinov, 1965) on the basis of its reliability, though it does not produce an appreciable error until very low supersaturations ($<0.1\%$) are employed (Twomey, 1967).

It is evident from the above that the thermal diffusion chamber has become a very useful and handy tool for studying different properties of the CCN. However, it is recognized (Twomey, 1963) that this chamber has some operational and fabrication problems which need to be carefully examined before its measurements can be relied upon. Very recently, Braham (private communication) pointed out to us that one of his graduate students, Mr. James W. Fitzgerald, had shown that the transient behavior of this chamber may be a source of considerable error in some of the CCN measurements.

In this article we present the explicit solution of the equations governing the transient behavior of the chamber. Fitzgerald's (1970) conclusion that the

chamber may experience some short-lived peak supersaturations which are several times higher than the maximum steady-state supersaturation if the incoming sample is saturated at a temperature less than that of the hot top plate, is also verified. We feel, however, that even if the incoming sample is introduced at the top plate temperature, these detrimental transient supersaturation peaks may arise if the sample is not drawn into the chamber in laminar flow. The transient supersaturation profiles may be readily calculated for any chamber under a given set of operating conditions from the plots of the dimensionless function presented in the following section. The guidelines for selecting optimum operating and design parameters for a chamber are discussed. We have constructed a thermal diffusion chamber of a similar design as that of Kocmond and Jiusto (1967). Using this chamber, the CCN concentration at Rolla was measured as a function of time during May 1969 under changing weather conditions. The results of these preliminary investigations are also reported.

2. Theory of operation

Assuming one-dimensional diffusion and temperature fields in the chamber, and the vapor concentration diffusion and heat conduction as the only source, respectively, for these fields, the diffusion equations can be written (Amelin, 1967) as

$$\frac{\partial p}{\partial t} = D \frac{\partial^2 p}{\partial x^2}, \quad (1)$$

$$\frac{\partial T}{\partial t} = \kappa \frac{\partial^2 T}{\partial x^2}. \quad (2)$$

Here p is the partial vapor pressure, T the temperature, t time, D and κ are the vapor and heat diffusion coefficients, respectively (assumed temperature independent), and the x coordinate has been taken along the height of the chamber. Eq. (1) implies that the phenomenon of thermal diffusion does not contribute appreciably to the transfer of vapor molecules. This is a valid assumption as long as large temperature gradients are not employed (Mason *et al.*, 1966) which, of course, is the usual case for CCN counting applications since only supersaturations as high as a couple of per cent are customarily produced. However, for studying homogeneous nucleation this effect cannot be neglected (Katz and Ostermier, 1967). A similar argument holds for Eq. (2) where the heat transport due to the flux of molecules is assumed negligible.

Let us now consider a chamber of height h whose top plate is maintained at a temperature T_1 and the bottom at T_2 such that $T_1 > T_2$. The two plates are kept moist by suitably placing wetted filter papers so that the partial vapor pressures at the top and bottom plate are p_1 and p_2 , respectively, which correspond to the

equilibrium vapor pressure values at the two respective temperatures. In order to provide a well-defined initial condition, let us further assume that the incoming sample is at a temperature T_0 , which for the sake of generality has been taken different from T_1 and T_2 , and contains water vapor at pressure p_0 saturated at T_0 . The initial and boundary conditions for Eqs. (1) and (2) then become

$$p(x,0) = p_0, \quad p(0,t) = p_2, \quad p(h,t) = p_1, \quad (3)$$

$$T(x,0) = T_0, \quad T(0,t) = T_2, \quad T(h,t) = T_1. \quad (4)$$

Using the straightforward procedure of separation of variables and Fourier's theorem, solutions of Eq. (1) and (2) under conditions (3) and (4), respectively, can be expressed as

$$p(x,t) = \sum_{n=1}^{\infty} \left\{ \left[-\frac{2(p_2-p_0)}{n\pi} + \frac{2(p_1-p_0)}{n\pi}(-1)^n \right] \times \sin\left(\frac{n\pi x}{h}\right) \exp\left[-\left(\frac{n\pi}{h}\right)^2 Dt\right] \right\} + \left(\frac{p_1-p_2}{h}\right)x + p_2, \quad (5)$$

$$T(x,t) = \sum_{n=1}^{\infty} \left\{ \left[-\frac{2(T_2-T_0)}{n\pi} + \frac{2(T_1-T_0)}{n\pi}(-1)^n \right] \times \sin\left(\frac{n\pi x}{h}\right) \exp\left[-\left(\frac{n\pi}{h}\right)^2 \kappa t\right] \right\} + \left(\frac{T_1-T_2}{h}\right)x + T_2. \quad (6)$$

Eqs. (5) and (6) govern the distribution of vapor pressure and temperature, respectively, in the chamber until the steady state is reached. When time becomes sufficiently large for the chamber to establish the steady state, the first term in both these equations vanishes and the steady-state linear vapor and temperature profiles are thus generated, i.e.,

$$p = p_2 + \left(\frac{p_1-p_2}{h}\right)x, \quad (7)$$

$$T = T_2 + \left(\frac{T_1-T_2}{h}\right)x. \quad (8)$$

Eliminating the variable x in Eqs. (7) and (8), we may express (Amelin, 1967) the partial vapor pressure as a function of temperature as

$$p = \left(\frac{T-T_2}{T_1-T_2}\right)(p_1-p_2) + p_2. \quad (9)$$

Let us now define the supersaturation S as the ratio of the vapor pressure p in the gas phase at a temperature T to the saturated or equilibrium vapor pressure p_e above the plane surface of the same liquid at the same temperature, i.e.,

$$S = \frac{p}{p_e}. \quad (10)$$

Using Eq. (9), the steady-state supersaturation in the chamber can be written as

$$S = \left(\frac{T-T_2}{T_1-T_2}\right)\left(\frac{p_1-p_2}{p_e}\right) + \frac{p_2}{p_e}. \quad (11)$$

The maximum steady-state supersaturation S_m existing in the geometrical middle of the chamber may be obtained from Eq. (11) in the form

$$S_m = \frac{p_1+p_2}{2p_e(T_m)}, \quad (12)$$

where $p_e(T_m)$ represents the saturated vapor pressure at the mean temperature T_m .

a. Transient behavior of the chamber

The unsteady-state supersaturation profile in the chamber can be calculated with the help of Eqs. (5) and (6) knowing the temperatures of the top and bottom plates and the temperature and vapor pressure of the incoming sample. Let us consider below the two cases of practical interest.

CASE I. When the incoming sample is at the bottom plate temperature and saturated, i.e., $T_0 = T_2$, $p_0 = p_2$, Eqs. (5) and (6) can be combined into a dimensionless form as

$$\xi(x',t') = x' + \frac{2}{\pi} \sum_{n=1}^{\infty} \left[\frac{(-1)^n \sin(n\pi x')}{n \exp(n^2\pi^2 t')} \right], \quad (13)$$

where

$$x' = \frac{x}{h}, \quad (14)$$

$$\xi(x',t') \equiv \xi_p(x',t') = \frac{p(x,t) - p_2}{p_1 - p_2}, \quad \text{when } t' = \frac{Dt}{h^2}, \quad (15)$$

and

$$\xi(x',t') \equiv \xi_T(x',t') = \frac{T(x,t) - T_2}{T_1 - T_2}, \quad \text{when } t' = \frac{\kappa t}{h^2}. \quad (16)$$

CASE II. When the incoming sample is at the top plate temperature and saturated, i.e., $T_0 = T_1$, $p_0 = p_1$,

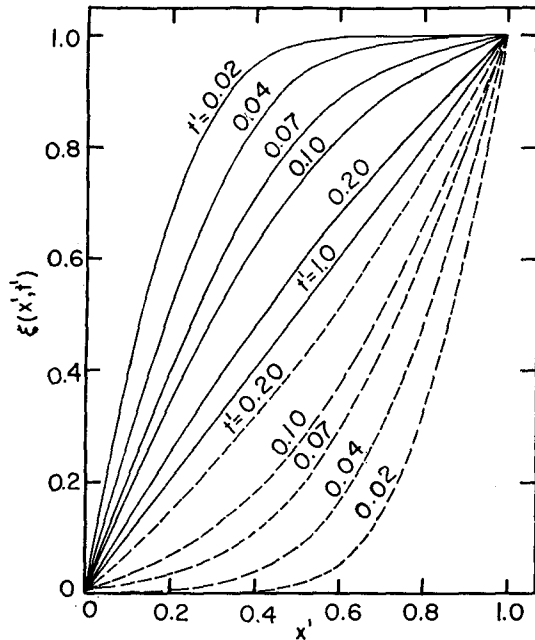


FIG. 1. Generalized plot of the dimensionless function $\xi(x', t')$ vs the scaled chamber height x' for different values of the dimensionless time variable t' . The dashed curves refer to case I where the incoming sample is saturated at the cold bottom plate temperature, while the continuous curves refer to case II where the incoming sample is saturated at the hot top plate temperature.

the dimensionless form of Eqs. (5) and (6) is

$$\xi(x', t') = x' + \frac{2}{\pi} \sum_{n=1}^{\infty} \left[\frac{\sin(n\pi x')}{n \exp(n^2 \pi^2 t')} \right]. \quad (17)$$

Fig. 1 shows $\xi(x', t')$ plotted as a function of t' . The dashed curves in this figure belong to case I while the continuous curves refer to case II. These plots can be used in conjunction with Eqs. (14)–(16) to generate the transient supersaturation profiles for any thermal diffusion chamber under given operating conditions. For all practical purposes, when t' attains the unit value, the profile of $\xi(x', t')$ becomes linear; this corresponds to the steady state. One may, therefore, also estimate from Eqs. (15) and (16) the time required for the vapor and temperature profiles to achieve steady states. Obviously, this time would be shortened for a shallower chamber.

Fitzgerald (1970) has shown that case I is detrimental to the chamber operation as transient peaks occur in the supersaturation profile which are higher than the maximum steady-state supersaturation obtained in the middle of the chamber. This is, however, true only if the incoming sample is saturated at the bottom plate temperature. The magnitude of these peaks certainly depends upon the relative humidity of the sample. This is evident from Fig. 2 where the supersaturation profiles are plotted for our chamber operating under case I corresponding to 50% relative humidity

of the sample. The temperature difference between the plates of the chamber of height 1.5 cm is assumed to be 10C while its bottom plate is at 21C. The values of D and κ at the mean temperature are taken as 0.258 and 0.219 $\text{cm}^2 \text{sec}^{-1}$, respectively. As $D > \kappa$, the vapor pressure profile increases faster toward the steady state than the temperature profile, but this effect is offset and dominated by the large difference between the vapor pressure of the incoming sample and its steady-state values in the chamber. This is what prevents the higher transient supersaturation peaks from occurring. Consequently, if one wants to operate the chamber under case I, one has to monitor and control the relative humidity of the sample; this is not likely to be convenient from a practical viewpoint.

b. Consideration of operational and design parameters

It is apparent from the above that prudent chamber operation would restrict the temperature or the relative humidity of the incoming sample. The most favorable condition seems to be the introduction of the sample into the chamber at the hot top plate temperature (Fitzgerald, 1970). Another operational problem then arises as how to introduce the sample so that it does not encounter a hotter moist surface. This, in turn, strongly dictates the design of the sample introducing device. The usual practice of venting in the sample through a small orifice or a valve does not seem to prevent turbulence. The turbulence invariably brings the incoming sample, which finally becomes quiescent between the chamber plates, in contact with the hotter moist surface thereby generating unpredictable transient supersaturation profiles. The existing literature reports chamber operation with the incoming sample at the hot top plate temperature (Twomey, 1963; Koc-

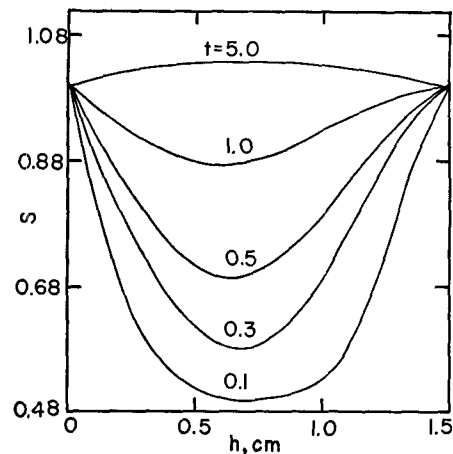


FIG. 2. Plot of the supersaturation S vs chamber height h at different times t (sec). The chamber of height 1.5 cm is assumed to have a temperature difference of 10C with the bottom plate maintained at 21C. The incoming sample is at 21C with relative humidity of 50%.

mond and Jiusto, 1967) and also at the cold bottom plate temperature (Radke and Hobbs, 1969a).

Apart from the sampling system, the chamber design also requires a careful consideration of the wall and vapor depletion effects. Twomey (1963) has pointed out that from both these considerations a shallower chamber is preferable, but this, in turn, limits the effective aperture of the illuminating optics if the photographic method is used for the nuclei counting. Our conclusion is that the chamber should be as shallow as possible and that the sample should be introduced in laminar flow at the temperature of the hot top plate. Under these conditions the peaking supersaturation phenomenon accompanying the transient behavior is eliminated, and the vapor depletion and wall effects are minimized.

3. Experimental results

The schematic of our thermal diffusion chamber is shown in Fig. 3. The design is very similar to that of Kocmond and Jiusto (1967, 1968). A ring of plexiglass about 10 cm in diameter and 1.5 cm in height is sandwiched between two porous stainless steel plates covered with filter papers. This ring has three glass windows, each 1 cm in diameter, two of which are used to receive and transmit a parallel 1 cm diameter beam of light from a suitably housed 100 W mercury arc, and the third is used to photograph the drops with a Nikon camera through a magnifying lens at right angles to the beam. The chamber is placed between the two 2.5 cm thick copper plates, each of which is heated or cooled as needed by four thermo-electric modules (Model 940 supplied by Borg-Warner Company). An aluminum heat sink with a fan is provided for each set of thermo-electrics to dissipate the unwanted heat. These thermo-electrics are energized by a transistorized power supply to produce any desired temperature gradient. The atmospheric sample is introduced into the chamber with the help of a vacuum connection through a 6 inch long, $\frac{1}{4}$ inch diameter, tygoon tubing which, in turn, is connected to one end of the $1\frac{1}{8}$ inch i.d., 8 ft long, copper pipe. The other end of this pipe is opened into the atmosphere, ~ 12 ft above the ground, extending ~ 6 ft beyond the building wall, while the remaining 2 ft length is kept at room temperature. The atmospheric aerosol is pulled through this pipe at a controlled flow rate by a small centrifugal fan. The sample does not come in contact with the fan before entering the chamber. During the measurements reported here the top plate was maintained at 24.9C and the bottom at 23.1C. The room temperature during these investigations was 23.5C which may be taken as the temperature of the test sample. The temperatures of the top and bottom plates were measured using a copper-constantan thermocouple before starting the CCN concentration measurements. This calibrated the operating temperature of the chamber in terms of the settings on the proportional temperature controller. The measurements

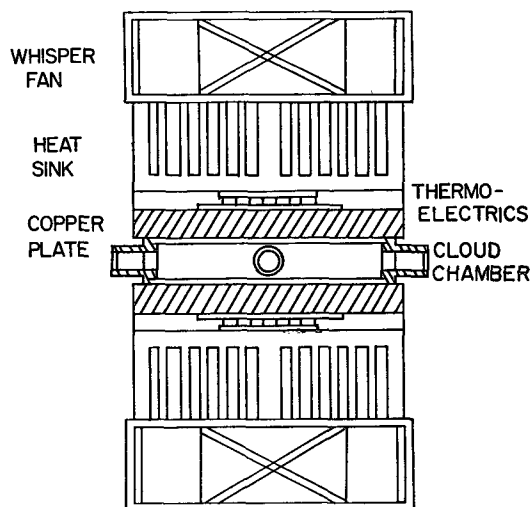


FIG. 3. A cross-sectional view of the thermal diffusion chamber.

were made during 22–25 May 1969 when the weather conditions changed from the worst on the morning of 22 May to the best at noon on 25 May as regards the visibility in the atmosphere.

Before presenting the results of our measurements let us point out that the design and the operation of our chamber as employed in these investigations suffer from two shortcomings. First, our method of venting in the sample was not free from turbulence and, second, we did not have any control on the relative humidity of the incoming sample. We therefore regard our present data as provisional until it is checked with a chamber that is devoid of above defects. Indeed, the present results are comparable to those of previous investigators. We are perfecting the design of our chamber so as to comply with the theoretical requirements discussed in the preceding section and hope to present the results of more extensive investigations in the near future.

The steady-state temperature, vapor pressure and supersaturation profiles for the operating conditions of our chamber are displayed in Fig. 4, which clearly exhibits how the concavity in the equilibrium vapor pressure (p_e) curve produces a maximum steady-state supersaturation of about 0.2% at the middle of the chamber. The concentration of the CCN per arbitrary unit volume activated at this supersaturation is shown in Fig. 5 as a function of the time (CDT) during four days of our investigation. A visual estimate of the visibility is also shown plotted in this figure which is qualitatively classified into four categories as poor, fair, good and excellent. The visibility was regarded as poor when it was too foggy to see the objects beyond one-quarter mile. A comparison of the CCN concentration on different days of investigation is presented in Table 1.

Some general trends in the CCN concentration are evident from Fig. 5. In the morning of 22 May 1969, the

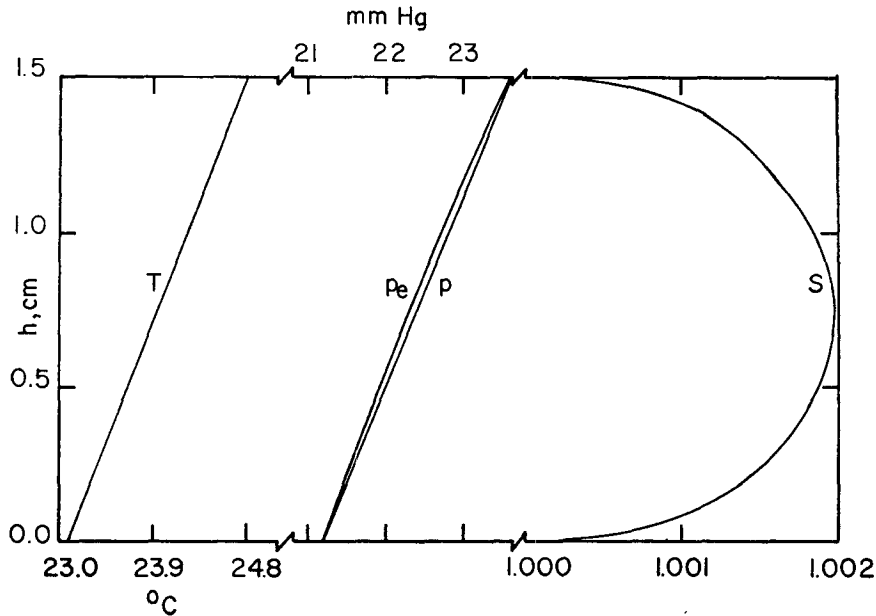


FIG. 4. Steady-state variation with the chamber height h of the temperature T , the partial vapor pressure p , the equilibrium vapor pressure p_e , and the supersaturation S .

clouds started building up and the visibility became poor. A slight improvement in the visibility was noticed during the afternoon though it remained mostly cloudy throughout the day. The variation in the CCN concentration recorded during this day typically reflects the unstable meteorological conditions. It went through a maximum in the afternoon and a minimum around midnight; throughout the afternoon and night, the wind was from the southeast at 10–15 mph. A good example, however, occurred on 23 May. The morning was mostly cloudy with very light easterly winds at 3 mph, and poor visibility. The CCN concentration started rising at noon when some light discontinuous drizzle occurred. Thereafter, the CCN concentration showed a steep fall from 201 to 79 within 4 hr. This reduction in the CCN concentration seems to be linked to the precipitation as has been pointed out by Radke and Hobbs (1969b).

On 24 May, the weather conditions remained relatively stable with cloudy skies and improved visibility

until 2100. The CCN concentration on the average was lower than that of the two preceding days. After 2100 the clouds started dissipating and within 2 hr the concentration rose from 57 to 224 though the sky was only partly cloudy. During this time the wind was light and northeasterly. This sudden rise in the CCN concentration may be taken as evidence in favor of a recent conclusion of Radke and Hobbs (1969b) that the dissipating clouds released particulates. During the morning of 25 May, the sky was cloudless, the visibility excellent and the wind very light. The average CCN concentration remained the lowest of all the four days. We hope to confirm some of these interesting conclusions through careful experimentation with our new chamber.

4. Conclusions

The following conclusions seem to be reasonable on the basis of our present study:

TABLE 1. Comparison of cloud condensation nuclei concentration (drop counts per arbitrary unit volume) activated at 0.2% supersaturation on different days of observation during 22–25 May 1969 at Rolla.

	Date							
	22 May		23 May		24 May		25 May	
	Time (CDT)	Concentration	Time (CDT)	Concentration	Time (CDT)	Concentration	Time (CDT)	Concentration
Median value	2200	119	2045	84	2000	44	1210	44
Maximum value	1630	171	1300	201	2300	224	2100	107
Minimum value	1230	64	2300	25	1900	23	1600	31

1) A brief review of the available literature on the thermal diffusion chamber reveals its wide and varied applications in studying problems concerning both homogeneous and heterogeneous nucleation. For investigating the various properties of cloud condensation nuclei, an understanding of which is so basic to weather modification and air pollution control, this chamber promises to emerge as an important tool provided its transient behavior is suitably handled.

2) Our analysis of the transient behavior confirms the conclusion of Fitzgerald (1970) that the transient supersaturations exceeding the steady-state peak value may arise if the incoming sample is saturated at a temperature less than that of the top plate. Such complications may be eliminated by introducing the sample into the chamber at the hot top plate temperature in a laminar flow. Consequently, the design and operation of the chamber should be modified to suit these requirements if dependable measurements are to be obtained. It is advantageous to know the transient supersaturation profiles in a chamber under a given set of operating conditions. These profiles may be readily computed from the dimensionless function plotted in Fig. 1.

3) Our measurements of CCN concentration under changing weather conditions using a conventional diffusion chamber confirm some of the short-term variations suggested by Radke and Hobbs (1969b). It is found that the precipitation reduces the CCN concentration, while dissipating clouds apparently enhance it.

4) It is felt that before accepting the thermal diffusion chamber as an undisputable and reliable CCN counter, further developmental work and careful experimental comparisons with suitable expansion cloud chambers are needed. The fact that some of the CCN concentration measurements by different cloud chambers show significant differences (Jiusto and Kocmond, 1968a) confirms this viewpoint.

Acknowledgments. We gladly acknowledge the stimulating discussions with Drs. John Carstens and J. F. Stampfer, Jr., during the tenure of this work. We are also thankful to Robert Sandler and Ronald Simmons for their help in the design and fabrication of the apparatus. It is our pleasure to sincerely acknowledge the invaluable suggestions and the editorial assistance extended to us by Prof. R. R. Braham, Jr. and Mr. J. W. Fitzgerald. We also feel indebted to Prof. Patrick Squires for pointing out the need for analyzing the transient behavior of the thermal diffusion chamber.

REFERENCES

Amelin, A. G., 1967: *Theory of Fog Condensation*. Israel Program for Scientific Translations, Jerusalem, p. 94.

Fitzgerald, J. W., 1970: Non-steady-state supersaturations in thermal diffusion chambers. *J. Atmos. Sci.*, **27**, 70-72.

Franck, J. P., and H. G. Hertz, 1956: Messung der kritischen Übersättigung von Dämpfen mit der Diffusionsnebelkammer. *Z. Physik*, **143**, 559-590.

Gagin, A., and B. Terliuc, 1968: A modified Wieland-Twomey thermal diffusion cloud nuclei counter. *J. Rech. Atmos.*, **2**, 73-77.

Hobbs, P. V., and L. F. Radke, 1969: Cloud condensation nuclei from a simulated forest fire. *Science*, **163**, 279-280.

Jiusto, J. E., 1966: Maritime concentrations of condensation nuclei. *J. Rech. Atmos.*, **2**, 245-250.

—, and W. C. Kocmond, 1968a: Note on cloud nucleus measurements in Lannemezan, France. *J. Rech. Atmos.*, **3**, 101-104.

—, and —, 1968b: Condensation on nonhygroscopic particles. *J. Rech. Atmos.*, **3**, 19-24.

Katz, J. L., and J. Ostermier, 1967: Diffusion cloud-chamber investigation of homogeneous nucleation. *J. Chem. Phys.*, **47**, 478-487.

Kocmond, W. C., and J. E. Jiusto, 1967: Project Fog Drops: Investigation of warm fog properties and fog modification concepts. Cornell Aero. Lab., Fourth Annual Rept., 56 pp.

—, and —, 1968: Investigation of warm fog properties and fog modification concepts. CAL Rept. RM-1788-P-17 (Clearing-house for Federal Scientific and Technical Information Springfield, Va., No. 22151).

—, and R. J. Pilie, 1969: Project Fog Drops: Investigation of warm fog properties and fog modification concepts. Cornell Aero. Lab. Quarterly Prog. Rept., No. RM-1788-P-22, 19 pp.

Laktinov, A. G., 1965: Determination of the concentration of cloud condensation nuclei. *Dokl. Akad. Nauk SSSR*, **165**, 1290-1293.

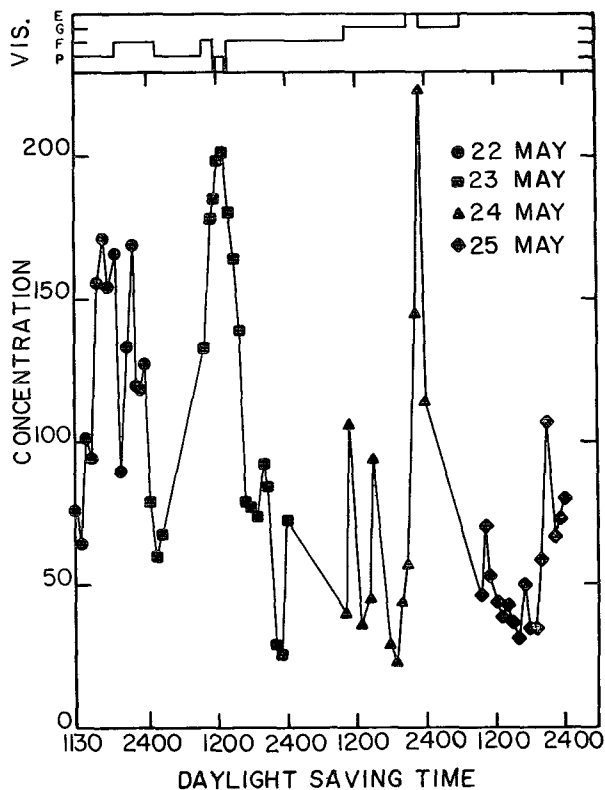


FIG. 5. Concentration (drop counts per arbitrary unit volume) of the CCN activated at 0.2% supersaturation as a function of time at Rolla, during the period 22-25 May 1969. A visual estimate of the visibility is also shown plotted, the letters having the following significance: P, poor; F, fair; G, good; E, excellent. Solid strips on the visibility plot mark the duration of very light drizzle.

- , 1967: On the connection between the condensation activity of irradiated nuclei and their sizes. *Izv. Atmos. Oceanic Phys.*, **3**, 25–33.
- , 1968: Photoelectric measurements of condensation cloud nuclei. *J. Rech. Atmos.*, **3**, 63–70.
- Langsdorf, A., 1936: A continuously sensitive cloud chamber. *Phys. Rev.*, **49**, 422.
- , 1939: A continuously sensitive diffusion cloud chamber. *Rev. Sci. Instr.*, **10**, 91–103.
- Mason, E. A., R. J. Munn and F. J. Smith, 1966: Thermal diffusion in gases. *Advances in Atomic and Molecular Physics*, Vol. 2, New York, Academic Press 33–91.
- Ohta, S., and E. Uchida, 1969: An automatic cloud condensation nucleus counter. *Proc. 7th Intern. Conf. on Condensation and Ice Nuclei*, Prague-Vienna, Abstract, p. 37.
- Radke, L. F., and P. V. Hobbs, 1969a: An automatic cloud condensation nuclei counter. *J. Appl. Meteor.*, **8**, 105–109.
- , and —, 1969b: Measurement of cloud condensation nuclei, light scattering coefficient, sodium-containing particles, and Aitken nuclei in the Olympic mountains of Washington. *J. Atmos. Sci.*, **26**, 281–288.
- Schiff, D., H. I. Schiff and P. R. Gendron, 1953: The use of a diffusion cloud chamber to characterize condensation nuclei. *Can. J. Chem.*, **31**, 1108–1117.
- Severynse, G. T., 1964: A portable cloud nuclei counter. *J. Rech. Atmos.*, **1**, 11–16.
- Squires, P., and S. Twomey, 1966: A comparison of cloud nucleus measurements over Central North America and the Caribbean Sea. *J. Atmos. Sci.*, **23**, 401–404.
- Storozhilova, A. I., 1961: A differential counter for condensation nuclei. *Sb. Issledovanie v Oblasti Poverkhnostnykh Sil* (Researches in the Field of Surface Forces), Moscow, Akad. Sci., USSR, 209–211.
- Twomey, S., 1959a: The nuclei of natural cloud formation. Part I: The chemical diffusion method and its application to atmospheric nuclei. *Geofis. Pura Appl.*, **43**, 227–242.
- , 1959b: The nuclei of natural cloud formation. Part II: The supersaturation in natural clouds and the variation of cloud droplet concentration. *Geofis. Pura Appl.*, **43**, 243–249.
- , 1960: On the nature and origin of natural cloud nuclei. *Bull. Obs. Puy de Dome*, **1**, 1–19.
- , 1963: Measurements of natural cloud nuclei. *J. Rech. Atmos.*, **1**, 101–105.
- , 1965: Size measurements of natural cloud nuclei. *J. Rech. Atmos.*, **2**, 113–119.
- , 1967: Remarks on the photographic counting of cloud nuclei. *J. Rech. Atmos.*, **3**, 85–90.
- , 1968: On the composition of cloud nuclei in the north-eastern United States. *J. Rech. Atmos.*, **3**, 281–285.
- , and G. T. Severynse, 1964: On the relation between sizes of particles and their ability to nucleate condensation of natural clouds. *J. Rech. Atmos.*, **1**, 81–85.
- , and J. Warner, 1967: Comparison of measurements of cloud droplets and cloud nuclei. *J. Atmos. Sci.*, **24**, 702–703.
- , and T. A. Wojciechowski, 1969: Observations of the geographical variation of cloud nuclei. *J. Atmos. Sci.*, **26**, 684–688.
- Vollrath, R. E., 1936: A continuously active cloud chamber. *Rev. Sci. Instr.*, **7**, 409–410.
- Volmer, M., 1939: *Kinetik der Phasenbildung*. Dresden and Leipzig, Steinkopff, p. 100.
- Warner, J., 1968: The supersaturation in natural clouds. *J. Rech. Atmos.*, **3**, 233–237.
- Wieland, W., 1956: Die Wasserdampfkondensation an natürlichem Aerosol bei geringen Übersättigungen. *Z. Angew. Math. Phys.*, **7**, 428–460.