Numerical Simulation of a Widely Used Cloud Nucleus Counter

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ABSTRACT

The performance of the conventional horizontal plate thermal diffusion chamber as a cloud nucleus counter was studied. Numerical calculations were performed in order to follow the simultaneous drop growth and sedimentation in the spatially and temporally nonuniform supersaturation field. The effect of nuclei distribution and smallest detectable drop size were investigated. The results indicate order of magnitude uncertainty in the count at 0.1% supersaturation, and a factor of 2 uncertainty at 1.0% supersaturation.

1. Introduction

The horizontal plate thermal diffusion chamber, invented by Langsdorf (1936), introduced to the cloud physics community by Wieland (1956), and developed for cloud physics use by Twomey (1963), has become an almost standard instrument for counting CCN. The lower limit of supersaturation at which this instrument should be used has been crudely estimated to be 0.1% (Twomey, 1967; Squires, 1972) and 0.2% (Sinnarwalla and Alofs, 1973). In recognition of increasing inaccuracy as the supersaturation is lowered, the lowest value used at the 1970 Fort Collins Workshop was 0.3%; however, no quantitative error estimates have been available. In order to obtain such error estimates, we performed a detailed numerical simulation of the instrument.

The growth of the water droplets in the chamber was treated by allowing them to fall at their terminal velocity (Stokes' law) and simultaneously grow in the spatially and temporally nonuniform environment produced in the chamber. The concentration n of nuclei with critical supersaturation S_c below S was assumed to be of the form

$$n = CS^K, \tag{1}$$

where C and K are constants. Various values of K between 0.15 and 2.0 were assumed.

The concentration of droplets was assumed sufficiently dilute so that they could be regarded as isolated insofar as both the calculations of fall and growth are concerned. This dilution was also assumed sufficient to neglect supersaturation decrements caused by vapor depletion and condensation heat release.

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2. The calculation of chamber supersaturation

For all calculations, the initial temperature of the sample was taken to be the warm plate (top plate) temperature, the initial relative humidity of the sample to be 90%, the average of the two plate temperatures to be 25°C, and the plate spacing to be 1 cm. The values used for all thermophysical properties are given in Table 1 of Sinnarwalla *et al.* (1975) as a function of temperature.

Even though the conventional relaxation times are about 0.5 s for the chamber herein described, the mid-plane transient supersaturation can take several times this long to reach 63% of its final mid-plane value. This duration will often turn out to be a non-negligible fraction of the growth time; the transient regime was therefore included in the simulation. Transient supersaturation profiles within the chamber were calculated using the series solutions of Fitzgerald (1970) and Saxena *et al.* (1970). The former is expressed in terms of complementary error functions and converges rapidly for short times; the latter, in terms of sines, provides better convergence for long times. Vapor pressure, rather than density, was used as the diffusant variable in accordance with the suggestion of Katz and Mirable (1975). Both solutions were found to satisfy the obvious requirement that they give identical numerical results.

3. Calculation of drop growth

The standard meteorological theory, in the form described by Carstens *et al.* (1974), was enlisted to follow the growth process of the solution droplets. Throughout the calculations, the thermal accommodation coefficient and the condensation coefficient were assumed to be 1.0 and 0.03, respectively, in accord with recent measurements (Chodes *et al.*, 1974; Sinnarwalla

et al., 1975). The vertical velocity of the drops was taken to be the terminal gravitational settling of the drops as given by Stokes' law (Fuchs, 1964).

The calculations followed the following format: For a particular set of plate temperatures, a nucleus with a specific value of S_c was located initially at some distance z_0 from the top plate. Numerical calculations were then performed in order to solve the growth equation and the Stokes' equation simultaneously. In order to provide a check on the solution of these two equations, two separate computer programs were prepared. One utilized Hamming's modified predictor corrector method.[3] The other utilized straightforward central differencing techniques. For all cases considered, agreement between the two was within 3% on either drop radius r or drop position z measured from the top plate. In the majority of the cases the agreement was better than 1%.

Using the above procedure drop radius was calculated as a function of time, for specific combinations of z_0 and S_c, and specific plate temperatures. Three sets of plate temperatures were chosen, corresponding to chamber operating supersaturations S_0 of 0.1, 0.3 and 1.0%. By S_0 we mean the final value of supersaturation produced at the mid-plane of the chamber. For each value of S_0, six values of z_0 were tested, *viz.*, 0, 0.1, 0.2, 0.3, 0.4, 0.5 and 0.6 cm. For each value of z_0, about 20 values of $S_c < S_0$ were tested. The values were rather closely spaced near $S_c = S_0$, and farther spaced for $S_c \ll S_0$. The results were printed out for each second interval of time.

4. Calculations of instrument response

In the chamber used by Twomey (1963) and many others, the drops are counted photographically, by illuminating the central portion of the chamber, and taking several pictures as time progresses. The time when the maximum number of drops appears on the photograph is called the time of the maximum cloud. The number of drops on the photograph at this time, divided by the viewed volume of the chamber, is taken as the concentration of nuclei active at S_0.

To evaluate the chamber performance from our calculations, we assumed that the central 2 mm region of the chamber is illuminated. Thus the viewed volume begins at $z=0.4$ cm and ends at $z=0.6$ cm. The other parameter of interest is the smallest size drop recorded on the film. We call this the detector limit r_0, and considered the three cases $r_0=0.5$, 1.0 and 1.5 μm radius.

Let S_{c_1} denote the least upper bound of S_c for nuclei which fall out of the view volume. Thus S_{c_1} is a function of time t and z_0, the initial nucleus position.

Similarly let S_{c_2} denote the least upper bound of S_c for nuclei which fall in or remain in the view volume and which reach the detector size limit. Thus S_{c_2} is a function of t, z_0 and r_0.

[3]IBM system/360 scientific subroutine package: IBM Application Program Manual, 1968.

Also let h be the unit step function such that $h=0$ for $S_{c_2} < S_{c_1}$ and $h=1$ for $S_{c_2} \leqslant S_{c_1}$.

Then recalling that K is the exponent in the assumed nucleus distribution (1), the count n can be expressed as

$$n = C \int_{z_0=0}^{z_0=0.6\text{ cm}} (S_{c_2}^K - S_{c_1}^K) h \, dz_0. \tag{2}$$

The correct count n_0 is given by

$$n_0 = C \int_{z_0=0.4\text{ cm}}^{z_0=0.6\text{ cm}} S_0^K \, dz_0, \tag{3}$$

where S_0 is the steady-state mid-plane supersaturation. In both (2) and (3) C is the same constant.

To find values of S_{c_2} and S_{c_1} from the calculations described in Section 3, two series of graphs were made. These graphs allowed interpolation between the data points provided by the calculations. Each graph was for a particular instant of time. In the first series of graphs, lines of constant z_0 were plotted with S_c on the abscissa and drop position z on the ordinate. Lines of constant drop radius were also drawn on these graphs. From such graphs, we obtained values of S_{c_2} and S_{c_1} for a finite number of values of z_0 between 0 and 0.6 cm. Lines of constant S_{c_2} and S_{c_1} were then plotted on a second series of graphs, with S_c on the abscissa and z_0 on the ordinate. This second series of graphs was mainly necessary to interpolate between $z_0=0.5$ cm and $z_0=0.6$ cm. Since a different graph was made for each instant of time, the number of graphs was rather large (about 40). Each graph in the first series involved plotting more than 100 data points so the human effort was considerable. Since the shape of the curves varied considerably from one graph to the next, there is no point in showing any of them without showing a large number of them. In the time region near the maximum cloud, the graphs were made with 1 s intervals between them.

Once values of S_{c_2} and S_{c_1} could be read from the above graphs for any z_0, the integral in (2) was approximated by finite difference using a summation of 12 terms, with $\Delta z_0=0.05$ cm. The value of S_{c_2} and S_{c_1} was recorded in a table for the center of each of these Δz_0 intervals. Then values of $S_{c_2}^K$ and $S_{c_1}^K$ for $K=0.15$, 0.5, 1.0 and 2.0 were calculated on a 10-digit electronic pocket calculator, entered in the table, and then added to obtain the count. In a few cases 24 terms rather than 12 were used, and very little difference in the result was found. In view of the accuracy with which the graphs could be read, and the accuracy of the finite-difference integration of (2), we estimate the overall error limit of the computed count to be $\pm 0.05 n_0$ or $\pm 0.2n$, whichever is greater.

5. Results

Fig. 1 shows the results for a chamber operating with a supersaturation of 0.1%. The ordinate is the ratio of

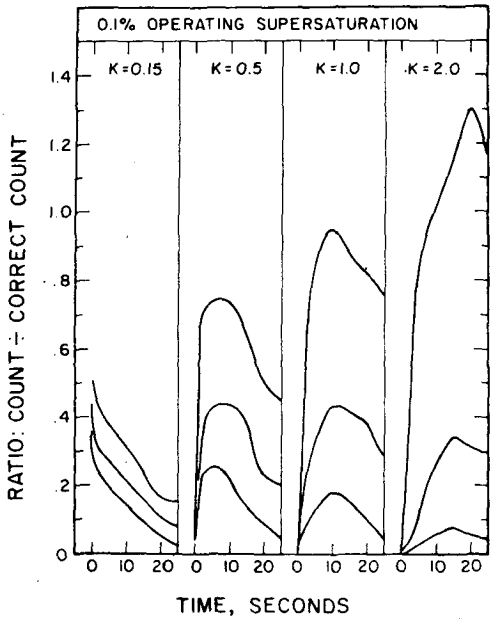

FIG. 1. Response at 0.1% supersaturation, for detector radius of 0.5 μm (top curve), 1.0 μm, and 1.5 μm: K=exponent in nuclei spectrum.

the counted nuclei divided by the correct count. The abscissa is the time since venting in the sample and sealing the chamber. The three curves in each panel are for different detector limits: the top curve for 0.5 μm radius, the middle for 1.0 μm radius, and the bottom for 1.5 μm radius. The four panels are for the indicated values of K, the exponent in the assumed nuclei distribution.

It can be seen from Fig. 1 that at 0.1% operating supersaturation the performance of the chamber depends strongly on both the detector limit and the nuclei distribution. For $K=2$, only 8% of the correct count is obtained with a 1.5 μm detection limit; whereas, for a 0.5 μm detection limit, the count is 30% above the correct count. Also, the time at which the maximum cloud appears is highly variable. It decreases as K decreases and as the detector limit decreases. Indeed, for $K=0.15$, the maximum count is at zero time, because of the relative importance of nuclei having large equilibrium radius at 90% relative humidity.

Fig. 2 shows the results at 0.3% operating supersaturation. Here the assumed nuclei distribution is not as important as at $S=0.1\%$, at least in the range $0.5<K<2$. However, the detector limit still influences the count strongly. It should be mentioned here that Twomey (1967) estimated the detector limit in his instrument to be between 0.5 and 1.0 μm radius. Chambers with less intense light sources would be expected to have higher detector limits, which may explain why the chamber at The University of Chicago was reported by Braham (1974) to give counts 50% lower than the chamber designed by Twomey at the

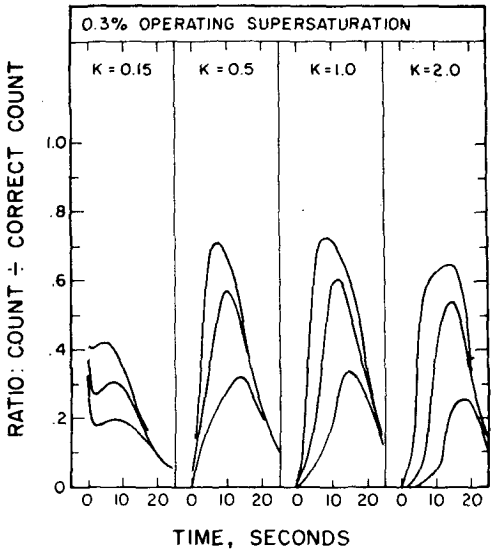

FIG. 2. As in Fig. 1 except for 0.3% supersaturation.

Naval Research Laboratory. Moreover, the detector limit undoubtedly varies considerably over the viewed volume of the chamber, both because the illumination beam is spatially nonuniform and because the scattering angle of the collected light varies with position in the view volume. Hence, for photographic counting, a detector limit uncertainty from 0.5 to 1.5 μm radius is not overly pessimistic.

Fig. 3 shows results at 1.0% operating supersaturation. For nuclei distributions with $0.5<K<2$, the count is 75 to 90% of the correct count, including the effect of detector limit uncertainty. However, for $K=0.15$, the count is as low as 50% of the correct count. Twomey and Wojciechowski (1969) report that

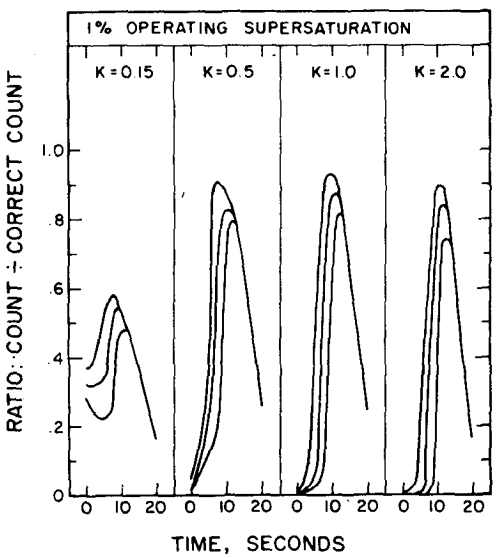

FIG. 3. As in Fig. 2 except for 1.0% supersaturation.

K values of 0.2 occur with a frequency of about 10% over land and sea. A review of Figs. 1, 2 and 3, however, will show that the horizontal chamber tends to overestimate K and thus that values of K as low as 0.15 should be expected to occur with significant frequency in the atmosphere. Thus, the horizontal chamber gives as much as 50% uncertainty at 1% supersaturation even when natural aerosols are used. When K is outside of the range tested, as is likely for artificial aerosols, the uncertainty is still greater.

In summary, our evaluation of the horizontal plate chamber indicates uncertainties of between 10 and 1000%, depending on the nuclei distribution, the detector limit, and the operating supersaturation of the chamber. Even at 1% supersaturation, errors of 50% are indicated. Moreover, variations in the initial sample humidity and in condensation coefficient, which our evaluation did not take into consideration, will add additional uncertainties.

6. Discussion

We have found rather large errors to be likely for the most widely used CCN counter. Let us now review some of the measurements which have been made with this instrument, and the conclusions made from these measurements. Fortunately, most of the conclusions do not rely upon the correctness of the absolute count, but rather are based upon examining the ratio of the count for one air parcel to the count for another air parcel. Thus numerous measurements indicated that there generally are more CCN in continental air masses than in marine (Twomey, 1959a, 1963; Squires and Twomey, 1966; Jiusto, 1966; Twomey and Wojciechowski, 1969); or downwind of St. Louis than upwind (Fitzgerald and Spyers-Duran, 1973). Because of the qualitative nature of this conclusion, the instrument errors we describe do not alter it.

There have been many quasi-simultaneous measurements made of CCN spectra below cloud base, droplet spectra within a cloud, and, sometimes, updraft speed (Twomey and Squires, 1959; Squires and Twomey, 1961; Twomey and Warner, 1967; Warner, 1968, 1969; Fitzgerald and Spyers-Duran, 1973). These measurements suggest that higher colloidal stability generally accompanies a higher CCN concentration, and this qualitative observation is likewise not influenced by the instrument errors we have found.

As to the quantitative aspects of the above measurements, they generally were compared with theory *via* the Twomey formula (1959b) which relates the CCN spectra, the updraft speed, and the resultant cloud droplet spectra. The best correlation gave factor of 2 discrepancies between observed and computed drop concentrations; however, Warner (1969) presents data with factor of 5 discrepancies, and states that most cloud traverses gave worse correlation than this. Most of these discrepancies are due to experimental difficulties of getting statistically representative data, but errors in the CCN counter are probably also significant.

For the past five years, Twomey and Davidson (1975) have made hourly measurements of CCN from a fixed ground station in Australia. So far, their data have shown a diurnal pattern but no discernible seasonal pattern or pattern for yearly averages. The conclusion concerning a diurnal pattern would not be affected by the instrument errors we have reported. With regard to any pattern of the yearly averages, however, more care must be taken, because the variation from year to year would probably be small. Inadvertent changes in the detector limit (smallest drop counted) might influence such a pattern significantly.

In summary, most of the conclusions based upon past measurements of CCN in the atmosphere will not be altered by the instrument errors we have reported. These instrument errors do, however, place limits on the type of measurements which can be done meaningfully. For future measurements which require a more accurate instrument several alternative instruments (Laktionov, 1972; Hudson and Squires, 1973; Sinnarwalla and Alofs, 1973) are available and should be considered.

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