A Vindication of the Twomey-Type Cloud Condensation Nucleus Counter

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(Manuscript received 5 August 1985, in final form 5 February 1986)

ABSTRACT

The performance of the Twomey-type cloud condensation nucleus (CCN) counter is evaluated by a numerical simulation and found to be better than that predicted in an earlier study. Due to a lack of data on CCN spectra at low supersaturations, the earlier study used extrapolations that subsequent measurements show are unrealistic for atmospheric CCN. The present study indicates that if the instrument has a 0.5 μm radius detector limit, the count is accurate to within 15% for operating supersaturations above 0.1% and for typical atmospheric aerosols.

1. Introduction

The performance of the Twomey-type CCN counter was evaluated by Alofs and Carstens (1976; henceforth denoted AC76). At that time little information was available on atmospheric CCN in the supersaturation (S) regime below 0.1%. Subsequently, measurements down to $S = 0.01\%$ have become available (Alofs and Liu, 1981). The CCN spectra assumed in AC76 were extrapolations in the low S regime, and such extrapolations turn out to be unrealistic for atmospheric CCN. The purpose of the present investigation is to perform similar computations as in AC76, but using CCN distributions more typical of the atmosphere. The effects of changing two other parameters, the initial relative humidity and the condensation coefficient, are also investigated.

In accord with the data reviewed or reported by Alofs and Liu (1981), the CCN spectra is represented by the following two-slope power law:

$$N = C_c S^{K_c}, \quad S > S^*, \tag{1}$$

$$N = C_f S^{K_c}, \quad S < S^*, \tag{2}$$

where N is the concentration of nuclei with critical supersaturation (S_c) below S and C_c, K_c, C_f, K_f, and S^* are constants. The subscripts in these constants have mnemonic value in that nuclei with $S_c < S^*$ have been given the name fog nuclei (Hudson, 1980), while nuclei with $S_c < 1\%$ have for decades been called cloud condensation nuclei. The value of S^* will be called the crossover supersaturation, because on a log–log plot of N versus S the two straight lines representing the spectrum cross at this S and the slope changes abruptly there.

In AC76 the initial relative humidity of the sample (Φ_0) was taken to be 90%, which simplified the calculations because the maximum molality of the solution droplets was then sufficiently low enough that Raoult's law could be applied. In the present investigation Φ_0 is taken to be 50%. The CCN are assumed to be pure ammonium sulfate, and the effect of the solute in lowering the vapor pressure is computed using the tabulated values of practical osmotic coefficients given by Robinson and Stokes (1959). For growth rate computations in the regime between dry salt particle and saturated solution droplet (with no remaining undissolved salt), the dissolution at the liquid–solid interface is assumed to be rapid compared to the condensation rate of water vapor.

In AC76 the condensation coefficient (β) was taken to be 0.03. The computations in this investigation are performed at two values of β, namely, 1.0 and 0.03. It turns out that these two values of β do not give rise to much difference in the indicated performance of the instrument. Also, the computations show that $\Phi_0 = 50\%$ gives nearly the same performance as $\Phi_0 = 90\%$. The most significant change from AC76 turns out to be the assumption of a two-slope CCN spectrum.

With the exception of the differences described above and some minor differences which are noted later, the assumptions, methods, and parameter values are the same as in AC76 and therefore will not be specified entirely here. The following assumptions are reiterated, however, because they are crucial to understanding the results. The drops are counted photographically by illuminating a region exactly midway between the hot and cold plates of the thermal diffusion cloud chamber. The plate spacing is 1.0 cm and the height of the illuminated region is 2 mm, as in AC76. The radius of the smallest droplet detected is called the detector limit and is denoted r_0. All droplets in the illuminated region are counted if they are larger than the detector limit. Photographs are taken in rapid succession after venting

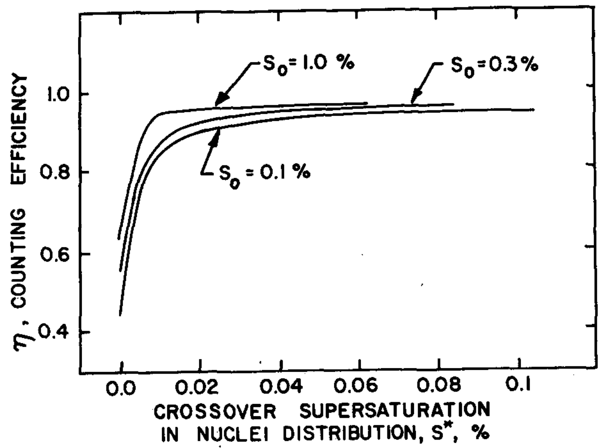

FIG. 1. The effect of S^* for the case r_0 = 0.5 μm, β = 1.0, Φ_0 = 50%, K_c = 0.15, and K_f = 1.0.

in the sample, and the time when the maximum number of drops appears on the photograph is called the time of maximum cloud. The number of drops on the photographs at this time, divided by the volume viewed on the photographs, is taken as the concentration of nuclei active at S_0, where S_0 is the steady state value of the supersaturation at the midplane of the chamber.

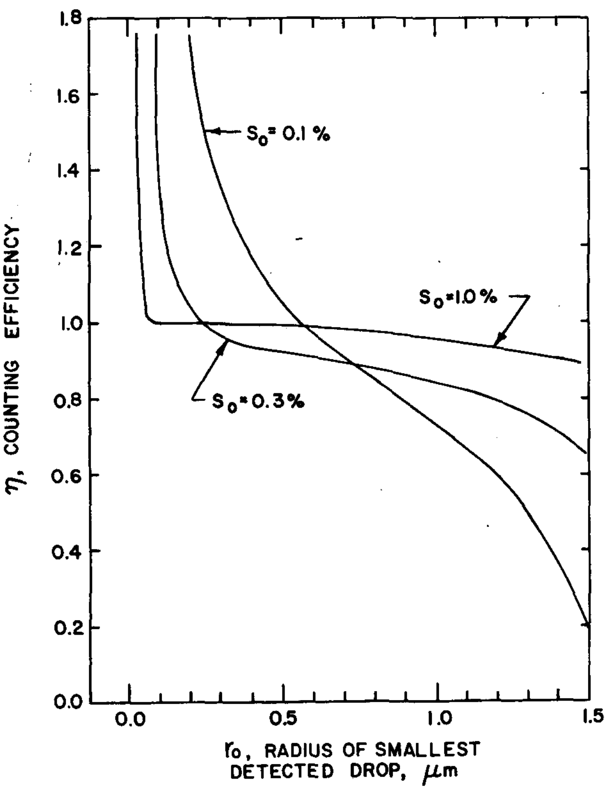

FIG. 2. The effect of r_0 for the case β = 1.0, Φ_0 = 50%, and the case 2 nucleus distribution (S^* = 0.0483%, K_c = 0.643, K_f = 3.85).

2. The crossover supersaturation S^*

From measurements at $S > 0.1\%$ made in the atmosphere over the past twenty years, values of K_c are known to range from 0.15 to 2.0 (Twomey and Wojciechowski, 1969). Measurements for $S < 0.1\%$ are more recent and more sparse. A survey of Alofs and Liu (1981) shows K_f values ranging from about 1.0 to 4.0, and S^* values from 0.025 to 0.2%. The values of C_c and C_f do not influence this investigation as long as they are consistent, such that (1) and (2) give the same N at S^*. This is because the concentration of droplets is assumed sufficiently dilute (Hudson and Squires, 1973) so that they can be regarded as isolated insofar as both sedimentation and growth are concerned.

Let η denote counting efficiency of the instrument, that is, the ratio of the measured N to the actual N. Figure 1 shows how S^* influences η, with other parameters held constant at the values indicated in the figure caption. The value chosen for K_c is 0.15, because in AC76 the performance of the instrument was very poor for a nuclei distribution of K = 0.15, where K denotes

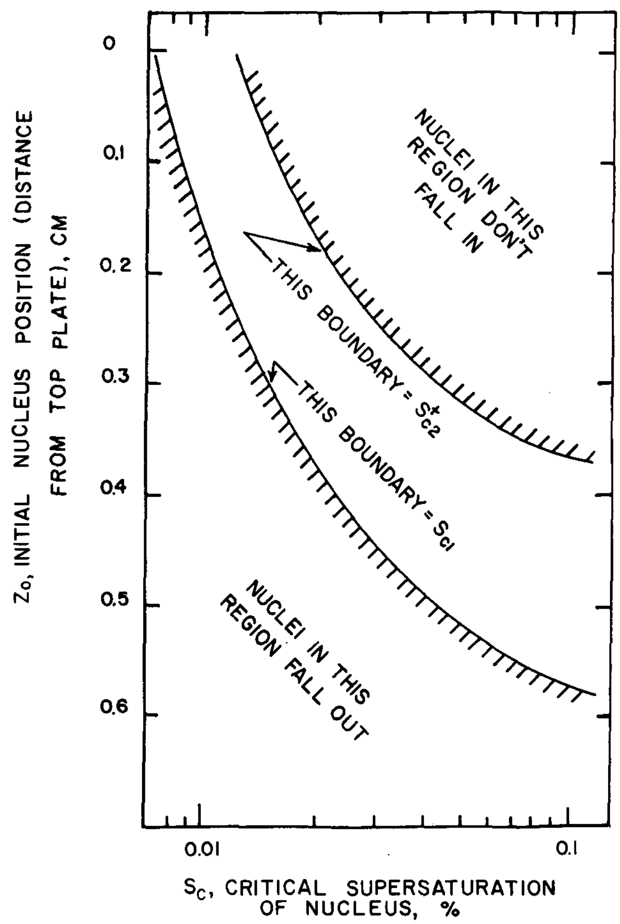

FIG. 3. Illustration of the S_{c1} and S_{c2}^+ boundaries. (Nuclei in these boundaries are in the illuminated region.)

the exponent in the single slope power law assumed in AC76. Figure 1 shows low values of counting efficiency at $S^* = 0$, in accord with the results of AC76. However, for S^* above 0.01%, Fig. 1 shows high counting efficiencies. Since the minimum S^* measured in the atmosphere is 0.025% (measured by Laktionov; see review by Alofs and Liu, 1981), we conclude that the assumed CCN distributions in AC76 were unrealistic for typical atmospheric conditions, and this gave rise to overly pessimistic interpretations of the results.

3. The detector limit (r_0)

The influence of r_0 on counting efficiency is shown in Fig. 2 for the indicated values of the other parameters. The CCN distribution is for the case $K_c = 0.643$, $K_f = 3.85$, and $S^* = 0.0483\%$. This distribution is the average distribution measured by Alofs and Liu (1981) during a 10-month atmospheric monitoring program at Rolla, Missouri. From now on this CCN distribution will be called case 2 and that of Fig. 1 for $S^* = 0.0483\%$ will be called case 1. It can be seen from Fig. 2 that for $S_0 = 1.0\%$, the counting efficiency is independent of r_0 over a large range of r_0. For $S_0 = 0.3\%$, the counting efficiency depends more strongly on detector limit, being 0.92 at $r_0 = 0.5$ μm and 0.73 at $r_0 = 1.0$ μm. Finally, at the lowest operating supersaturation studied ($S_0 = 0.1\%$), the performance of the instrument can be seen to depend critically on the value of r_0. AC76 also indicated that r_0 was important.

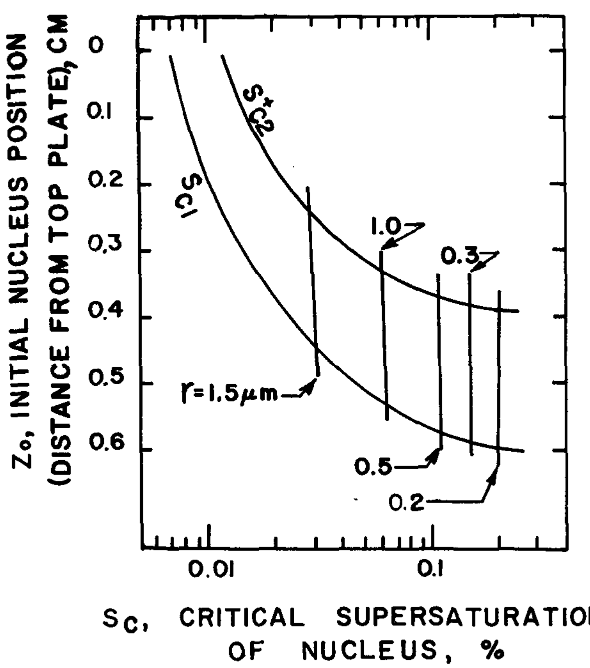

FIG. 4. S_{c1} and S_{c2} for $S_0 = 0.1\%$, $t = 10$ s, $\beta = 1.0$, and $\Phi_0 = 50\%$. (A line of constant drop radius, r forms part of the S_{c2} boundary for a detector limit equal to that drop radius.)

TABLE 1. Values of S_{c1} and S_{c2} for the case $S_0 = 0.1\%$, $r_0 = 0.5$ μm, $\Phi_0 = 50\%$, $\beta = 50\%$, and the indicated times. Here, z_0 is the initial nucleus position, expressed as the distance from the top plate in centimeters.

		$t = 9$ s		$t = 10$ s	
Row	z_0 (cm)	S_{c1} (%)	S_{c2} (%)	S_{c1} (%)	S_{c2} (%)
1	0.0125	0.00618	0.0102	0.0076	0.0124
2	0.0375	0.00622	0.0106	0.0077	0.0127
3	0.0625	0.0064	0.0113	0.0079	0.0135
4	0.0875	0.0065	0.0118	0.0082	0.0145
5	0.1125	0.0066	0.0128	0.0086	0.0156
6	0.1375	0.0068	0.0138	0.009	0.017
7	0.1625	0.0070	0.0154	0.0096	0.019
8	0.1875	0.0072	0.0172	0.0104	0.0215
9	0.2125	0.0076	0.0198	0.0112	0.0245
10	0.2375	0.0082	0.0235	0.0120	0.029
11	0.2625	0.0090	0.0280	0.0130	0.035
12	0.2875	0.0100	0.034	0.0145	0.041
13	0.3125	0.0110	0.043	0.016	0.050
14	0.3375	0.0126	0.058	0.0178	0.064
15	0.3625	0.0145	0.079	0.0198	0.083
16	0.3875	0.0170	0.108	0.0225	0.11
17	0.4125	0.0210	0.108	0.026	0.11
18	0.4375	0.024	0.108	0.0305	0.11
19	0.4625	0.029	0.108	0.0360	0.11
20	0.4875	0.0355	0.108	0.0430	0.11
21	0.5125	0.0440	0.108	0.0510	0.11
22	0.5375	0.0570	0.108	0.0650	0.11
23	0.5625	0.0790	0.108	0.0850	0.11
24	0.5875	0.108	0.108	0.11	0.11

Consider now the available data concerning r_0 values in Twomey-type CCN counters. Unfortunately, these data are quite sparse. The only published data we are aware of are by Twomey (1967). He used two techniques to measure r_0. One technique gave $r_0 = 0.5$ μm; the other gave 1.0 μm. Twomey rendered no judgment as to which technique was more reliable; he instead indicated that r_0 ranged from 0.5 to 1.0 μm in his instrument. Figure 2 indicates that a detector limit of 0.5 μm would give considerably better instrument performance than 1.0 μm. It therefore would be worthwhile to measure r_0 in individual Twomey-type CCN counters. Monodisperse aerosols generated with an electrostatic classifier (Liu and Pui, 1974) could provide accurate means to produce water drops of known stable size, by allowing salt nuclei to reach their equilibrium size at 100% relative humidity in the particular Twomey-type counter under study.

4. Sedimentation

Figure 3 illustrates the process of sedimentation in the chamber. The abscissa is the critical supersaturation of a nucleus. The ordinate is z_0, defined as the initial position of a nucleus, i.e., distance in centimeters from the top plate at time zero. Time (t) begins immediately after the sample vent-in process ceases. The two boundaries in Fig. 3 are denoted S_{c1} and S_{c2}^+. Here S_{c1} is defined as the least upper bound of S_c for nuclei that fall out of the illuminated region, and S_{c2}^+ is the greatest lower bound of S_c for nuclei that do not fall into the illuminated region. Thus at any particular time, only nuclei with $S_{c1} < S_c < S_{c2}^+$ are located in the illuminated region.

Let S_{c2} be the greatest lower bound of S_c for nuclei which have grown larger than the detector limit and which have $S_c < S_{c2}^+$. Figure 4 includes lines of constant droplet radius (r). Thus if $r_0 = 0.5$ μm, the S_{c2} line in Fig. 4 is congruent with the $r = 0.5$ μm line below the intersection point with the S_{c2}^+ line. Above this intersection point, the S_{c2} line is congruent with the S_{c2}^+ line. Now that S_{c2} is defined, we want to give the following addendum to the definition of S_{c1}: whenever the previous definition of S_{c1} gives $S_{c1} > S_{c2}$, define S_{c1} to equal S_{c2}. Then the number of droplets, n, counted on the photograph is given by the following integral:

$$n = \int_{z_0=0}^{z_0=0.6\text{ cm}} [N(S_{c2}) - N(S_{c1})]dz_0 \qquad (3)$$

where the horizontal cross-sectional area of the illuminated region has been assumed to be unity.

Table 1 gives values of S_{c1} and S_{c2} for an operating supersaturation of 0.1%, a detector limit of 0.5 μm, and two different values of time. Note that the S_{c1} at $z_0 = 0.4125$ cm is nearly the same as the value of S_{c2} at $z_0 = 0.2125$ cm. A similar effect can be seen in Table 1 at other pairs of z_0 values differing by 0.2 cm, the height of the illuminated layer. This is especially true for the larger values of z_0. The physical explanation is that the number of nuclei which fall out of the illuminated region is almost equal to the number which fall in. This suggests rearranging the information in Table 1 in the way shown in Table 2.

The second column in Table 2 assigns an error group number to collections of rows. The third column names pairs of S_c values. The fourth column gives the values of these pairs as read from Table 1 for 9 s. The fifth column gives the values at 10 s. Note that if the rows of expressions in the third column were added, the result would be $S_{c2} - S_{c1}$ (at each z_0 value), and a residual of $-8(S_0)$. Thus if each value of S_c in column 4 or 5 were replaced by $N(S_c)$ and the rows were added, the sum would equal the droplet count on the photograph minus the droplet count for an ideal instrument.

Table 3 shows the results of doing the above operation on Table 2 for two different nuclei distributions

TABLE 2. Rearrangement of Table 1 into error groups.

Row	Error group	$S_c(z_0)$ range of error group	Value of S_c range at $t = 9$ s	Value of S_c range at $t = 10$ s
1	I	$S_{c2}(0.4125)$–S_0	0.108–0.10	0.110–0.10
2	I	$S_{c2}(0.4375)$–S_0	0.108–0.10	0.110–0.10
3	I	$S_{c2}(0.4625)$–S_0	0.108–0.10	0.110–0.10
4	I	$S_{c2}(0.4875)$–S_0	0.108–0.10	0.110–0.10
5	I	$S_{c2}(0.5125)$–S_0	0.108–0.10	0.110–0.10
6	I	$S_{c2}(0.5375)$–S_0	0.108–0.10	0.110–0.10
7	I	$S_{c2}(0.5625)$–S_0	0.108–0.10	0.110–0.10
8	I	$S_{c2}(0.5875)$–S_0	0.108–0.10	0.110–0.10
9	II	$S_{c2}(0.2125)$–$S_{c1}(0.4125)$	0.0198–0.0210	0.0245–0.0260
10	II	$S_{c2}(0.2375)$–$S_{c1}(0.4375)$	0.0235–0.0240	0.0290–0.0305
11	II	$S_{c2}(0.2625)$–$S_{c1}(0.4625)$	0.0280–0.0290	0.0350–0.0360
12	II	$S_{c2}(0.2875)$–$S_{c1}(0.4875)$	0.0340–0.0355	0.0410–0.0430
13	II	$S_{c2}(0.3125)$–$S_{c1}(0.5125)$	0.0430–0.0440	0.0500–0.0510
14	II	$S_{c2}(0.3375)$–$S_{c1}(0.5375)$	0.0580–0.0570	0.0640–0.0650
15	II	$S_{c2}(0.3625)$–$S_{c1}(0.5625)$	0.0790–0.0790	0.0830–0.0850
16	II	$S_{c2}(0.3875)$–$S_{c1}(0.5875)$	0.108–0.108	0.110–0.110
17	III	$S_{c2}(0.0125)$–$S_{c1}(0.2125)$	0.0102–0.00760	0.0124–0.0112
18	III	$S_{c2}(0.0375)$–$S_{c1}(0.2375)$	0.0106–0.00820	0.0127–0.0120
19	III	$S_{c2}(0.0625)$–$S_{c1}(0.2625)$	0.0113–0.00900	0.0135–0.0130
20	III	$S_{c2}(0.0875)$–$S_{c1}(0.2875)$	0.0118–0.0100	0.0145–0.0145
21	III	$S_{c2}(0.1125)$–$S_{c1}(0.3125)$	0.0128–0.0110	0.0156–0.0160
22	III	$S_{c2}(0.1375)$–$S_{c1}(0.3375)$	0.0138–0.0126	0.0170–0.0178
23	III	$S_{c2}(0.1625)$–$S_{c1}(0.3625)$	0.0154–0.0145	0.0190–0.0198
24	III	$S_{c2}(0.1875)$–$S_{c1}(0.3875)$	0.0172–0.0170	0.0215–0.0225
25	IV	0–$S_{c1}(0.0125)$	0–0.00618	0–0.0076
26	IV	0–$S_{c1}(0.0375)$	0–0.00622	0–0.0077
27	IV	0–$S_{c1}(0.0625)$	0–0.00640	0–0.0079
28	IV	0–$S_{c1}(0.0875)$	0–0.00650	0–0.0082
29	IV	0–$S_{c1}(0.1125)$	0–0.00660	0–0.0086
30	IV	0–$S_{c1}(0.1375)$	0–0.00680	0–0.0090
31	IV	0–$S_{c1}(0.1625)$	0–0.00700	0–0.0096
32		0–$S_{c1}(0.1875)$	0–0.00720	0–0.0104

TABLE 3. Values of the error groups for two CCN distributions: case 1: $S^* = 0.0483\%$, $K_c = 0.15$, $K_f = 1.0$; case 2: $S^* = 0.0483\%$, $K_c = 0.643$, $K_f = 3.85$.

	Case 1		Case 2	
Error group	$t = 9$ s	$t = 10$ s	$t = 9$ s	$t = 10$ s
I	+1.160	+1.44	+5.072	+6.320
II	−1.206	−1.38	−1.074	−1.767
III	+2.92	−0.141	+0.129	−0.111
IV	−12.27	−16.01	−0.030	−0.088
Total error	−9.39	−16.09	+4.097	+4.345
Efficiency, n	0.906	0.839	1.041	1.043

(case 1 and case 2, defined previously). The value of C_c in the nuclei distributions has been chosen so that the correct count on the photographs is 100. The group sums in Table 3 correspond to the groups in the second column of Table 2.

The group I sum in Table 3 is the error in the count due to counting haze drops, i.e., drops grown on CCN with $S_c > S_0$. (The above statement applies only to this case. For $r_0 = 1.5$ μm, the group I error would be negative, the sign indicating undercount.) It can be seen from Table 3 that for the case 1 CCN distribution, the group I error is small compared to the total error, but for the case 2 CCN distribution, the group I error is predominant.

Let the 0.2 cm region immediately above the illuminated region be called region M and the 0.2 cm region above M be called T. (Mnenomic meaning of M is "middle" and of T "top.") The group II error is due to slight S_c mismatch between nuclei from region M that fall into (and perhaps also out of) the illuminated region, and the nuclei from the illuminated region that fall out of the illuminated region. The group III error is due to mismatch between the S_c of nuclei from region M that fall out of the illuminated region and the nuclei from region T that fall into (and perhaps also out of) the illuminated region. The group IV error is due to sedimentation out of the illuminated region of nuclei, which came from region T.

Table 3 shows that for the case 1 CCN distribution the predominant error is the group IV error. This error gets larger as time increases, and the time of maximum cloud is prior to 9 or 10 s. For the case 2 CCN distribution the time of maximum cloud is near 10 s and the Group IV error is very small, but the Group II error is significant. Table 3 shows how the major source for error shifts as the CCN distribution changes. Table 3 has allowed us to see in detail why Fig. 1 shows a counting efficiency below unity and Fig. 2 (for $r_0 = 0.5$ μm) shows a counting efficiency above unity.

5. Condensation coefficient and initial relative humidity

Figure 5 differs from Fig. 2 only in the value of the condensation coefficient (β). A comparison of these two figures indicates that a variation in β from 0.03 to 1.0 causes only slight changes in performance. The controversy concerning the correct value of β (see Wagner, 1982, versus Sinnarwalla et al., 1975) therefore has little bearing on the performance of these instruments. Figure 6 differs from Fig. 5 only in the value of the initial relative humidity of the sample (Φ_0). A comparison of these two figures indicates that a variation of Φ_0 from 50 to 90% causes only slight performance changes. Therefore, ambient relative humidity fluctuations in the range of 50 to 90% would not be expected to influence performance.

6. Conclusions

The predicted performance of the Twomey-type cloud condensation nucleus (CCN) counter is found to be better for "two-slope" atmospheric CCN spectra than for "single slope" CCN spectra. An earlier study (Alofs and Carstens, 1976) used only the single-slope spectra because at that time more complete data were not available. Subsequently, CCN measurements became available at lower supersaturations (below 0.1%), and these measurements gave rise to two-slope CCN spectra for characterizing atmospheric aerosols. Thus, the use of the single-slope spectra in the earlier study constituted an extrapolation of the CCN spectra into a region of low supersaturation for which there were

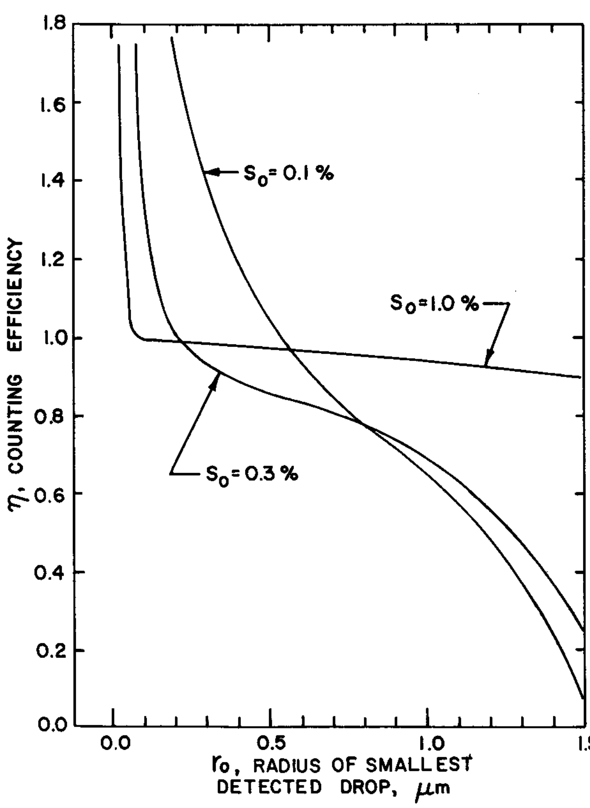

FIG. 5. As in Fig. 2, except $\beta = 0.03$.

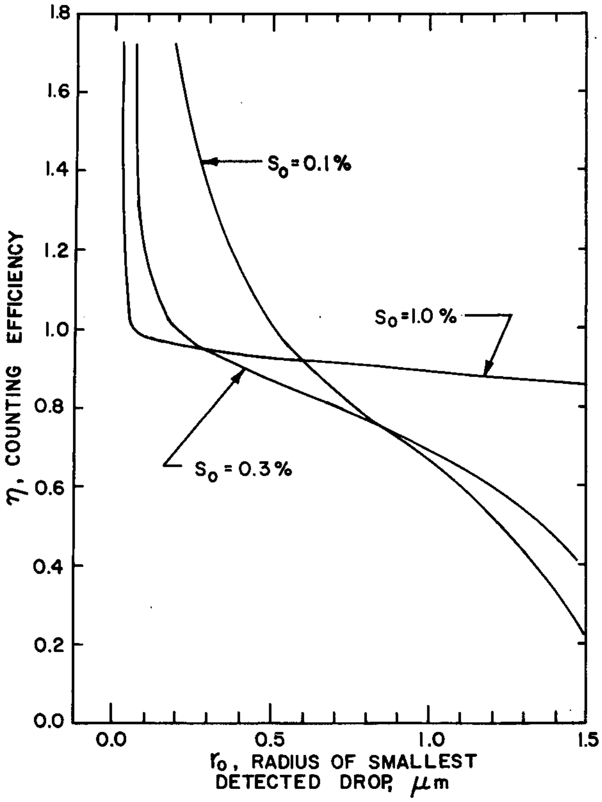

FIG. 6. As in Fig. 5, except Φ_0 = 90%.

no data. The present study shows that with more realistic atmospheric CCN spectra, the Twomey counter usually gives an accurate count if the radius of the smallest detectable droplet on the photographs is near 0.5 μm. The CCN distributions investigated in the present study showed uncertainties of about 15% in the measured nucleus concentration at 0.1% operating supersaturation, and lower uncertainties at higher supersaturation. The above conclusion, however, is only for the case where the detector limit is 0.5 μm. For higher value of detector limit, the uncertainty in the count increases.

A second conclusion is that at lower operating supersaturations, the performance of the instrument depends critically on the detector limit. The earlier study indicated this also. A detector limit of 0.5 μm radius appears to be about optimal. Thus, the authors recommend that users of a Twomey counter should determine the detector limit for their instrument and, if it differs significantly from 0.5 μm, change the design so as to achieve 0.5 μm. A recommendation of an experimental method to determine the detector limit is given at the end of section 3.

A third conclusion is that uncertainties about initial relative humidity (over the range 50 to 90%) or the condensation coefficient (over the range 0.03 to 1) are not of much importance. This adds to the generality of the performance computations done in this investigation and in AC76. It also means that one has some margin where one need not worry about changes in sample relative humidity, as when, for example, one takes a sample from the outdoors to the indoors through tubing that has temperature gradients.

A final conclusion is that for CCN spectra like those assumed in AC76, the performance would be as predicted in that study. The computations in AC76 are correct; only the interpretation that they represent typical atmospheric conditions is wrong. Since the performances predicted in AC76 were in some cases poor, this should serve as a warning that for some aerosols (perhaps artificially generated or atmospheric aerosols not well aged) the performance may be poor. This warning, of course, applies most strongly at the lower operating supersaturations.

Acknowledgments. This work was supported in part by the Atmospheric Science Section of the National Science Foundation, Grant NSF ATM79-24326, and constituted the Master of Science thesis of the second author.

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