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Atmospheric Measurements of CCN in the Supersaturation Range 0.013–0.681%

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ABSTRACT

Concentrations of cloud condensation nuclei in the atmosphere were measured at Rolla, Missouri, near ground level. A total of 432 spectra were measured over a 10-month period. The average concentration (N) of nuclei active at supersaturation, S, followed the two-stage power law:

$$N = 3990\ \text{cm}^{-3}\ S^{0.643},\quad 0.681\% \geq S \geq 0.0483\%$$

$$N = 6.62 \times 10^{7}\ \text{cm}^{-3}\ S^{3.85},\quad 0.013\% \leq S \leq 0.0483\%.$$

No consistent seasonal or diurnal pattern was observed. Possible implications for fogs and stratus clouds and for nuclei chemical composition are discussed.

1. Introduction

There is ample evidence that the supersaturation spectra of cloud condensation nuclei (CCN) influence the colloidal stability of cumulus clouds (Twomey and Squires, 1959; Twomey and Warner, 1967; Warner, 1969; and Fitzgerald and Spyers-Duran, 1973). Twomey (1959b) derived a simple equation relating the concentration (M) of cloud droplets to the updraft speed (U) and the CCN spectrum. The CCN spectrum was represented by the following power law:

$$N = CS^{k}, \tag{1}$$

where N is the concentration of CCN with critical supersaturation, S_c, below S, and C and k are constants. The Twomey equation indicates that for the low values of k(0.5–0.7) generally observed, M depends strongly on C and only weakly on U. Hence C, the CCN concentration parameter, dominates the microstructure of the cloud.

The above classical situation is based on the assumption that (1) is valid down to values of S well below S_{max}, the maximum supersaturation in the cloud. However, until recently, CCN measurements were made only for $S > 0.1\%$, due to limitations in the instrumentation for counting CCN (Squires, 1972; Alofs and Carstens, 1976). Recent advances in instrumentation (Alofs, 1978; Hudson, 1980) now allow making CCN measurements down to $S = 0.01\%$. The present paper reports CCN measurements made in the range $0.013\% \leq S \leq 0.681\%$, at Rolla, Missouri, during the period March–December, 1978. These measurements show that for $0.013\% \leq S \leq 0.05\%$, the average value of k is 3.85. With this value of k, and for clouds with $S_{max} \leq 0.05\%$, the Twomey equation would indicate that U dominates C in determining M. This has implications for stratus clouds and fogs, where the maximum supersaturation may be much lower than in cumulus clouds (Mason, 1960; Hudson, 1980; Gerber, 1981). Even for S_{max} somewhat above 0.05%, the effective value of k would be higher than the value at S_{max}, and hence U might dominate C.

2. Description of instrumentation

The instrument used for counting CCN has been described in detail (Alofs, 1978). It is a dual mode device. In the upper supersaturation range, $0.083\% \leq S \leq 0.681\%$, it operates as a continuous flow diffusion (CFD) cloud chamber. In the lower supersaturation range, $0.013\% < S < 0.122\%$, it operates as a haze cloud chamber using a technique first described by Laktionov (1972a).

The cloud chamber consists of two parallel, wet vertical plates 100 cm long in the vertical direction and 13 cm wide, with a 0.8 cm spacing between the plates. The sample air flows downward between the plates in a small diameter stream surrounded by filtered air. The sample flow is 8.5 cm^3 min^{-1}, determined by the pressure drop through a capillary tubing 0.25 mm diameter. This sample flow branches from a larger delivery flow of 5 L min^{-1} which in turn branches from a main duct flow of 300 L min^{-1}. This branching is to avoid losses of CCN in the process of bringing the sample from the atmosphere into the CCN counter.

The droplets formed on the CCN as they pass through the cloud chamber are counted and sized as

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they leave the chamber, using a Royco[1] Model 225 optical particle counter (OPC) with a five-channel pulse height analyzer (Royco Model 518). The rate of flow into the OPC is 0.35 to 1.0 L min^{-1}.

In the CFD mode, one plate is kept warmer than the other, which produces a supersaturation between the two wet plates. The warm plate is kept at 25°C, and the cold plate temperature is increased with time in steps, to produce supersaturations of 0.681, 0.436, 0.266, 0.170, 0.122 and 0.083%. Four minutes of data are taken at each supersaturation.

In the haze mode, both plates are kept at 25°C. The equilibrium radius, r_{100}, of the droplets at 100% relative humidity is measured with the OPC, and from this size, S_c is determined. Five minutes of data are taken at a filtered air flow rate of 0.35 L min^{-1} and another 5 min of data are taken at a filtered flow rate of 1.0 L min^{-1}. The data at 0.35 L min^{-1} is used for nuclei with $0.01\% < S_c < 0.03\%$, whereas the data at 1.0 L min^{-1} is used for nuclei with $0.03\% < S_c < 0.1\%$. The reason for this dual flow rate is that nuclei with small S_c have a big r_{100} and so require longer growth times, whereas nuclei with larger S_c have smaller r_{100} and hence must be brought through the OPC more quickly to avoid significant droplet evaporation.

The size thresholds of the Royco OPC were calibrated by using monodisperse NaCl aerosols while operating the cloud chamber in the haze mode. This procedure is described in a previous paper (Alofs *et al.*, 1979). One shortcoming of the Royco 225 should be discussed. There is a region of particle size where a given response corresponds to more than one particle size. During the 1978 sampling program herein described, the size thresholds of the Royco were set to avoid this region. Thus in the haze mode the supersaturation thresholds were set at 0.0133, 0.0267, 0.033, 0.044 and 0.122%. The gap from 0.044 to 0.122% was required in order to avoid the multivalue Royco response region. To help fill the gap, one of the CFD mode supersaturations was set at 0.083%.

The performance of the nucleus counter was tested at the 1980 Reno workshop[2] and found to be excellent. However, the question may be fairly raised as to whether verification of performance in 1980 is applicable to a measurement program made in 1978, especially since a different OPC (Climet[3] Model 210) was used at the Reno workshop. In response we state that no other changes were made in the instrument from 1978 to 1980. Also, it is noted that in 1978, studies with monodisperse NaCl aerosols were made using two cloud chambers, one equipped with a Royco 225 OPC, the other with a Climet 210 OPC. The nucleus concentrations measured by the two instruments agreed to within 10% accuracy in the CFD mode, and 20% accuracy in the haze mode, with the Royco thresholds set to avoid the multivalue Royco region. For these experiments the size thresholds of each OPC was calibrated using monodisperse NaCl aerosols. These experiments were published in part (Alofs *et al.*, 1979) but the concentration comparisons were not mentioned because the emphasis was on verifying the Köhler theory for NaCl.

3. Description of the site, the sampling duct and the schedule of sampling

The data were taken on the University of Missouri-Rolla campus, located in Rolla, Missouri. Rolla is a largely non-industrial town, with a population of 13 500. The closest large cities are St. Louis, located 167 km to the northeast; and Springfield, located 180 km to the southwest. The world's largest lead mining operation is located 100 km southeast of Rolla, and the refining operation associated with this district is known to produce sulfur dioxide emissions.

The air sampling duct protruded 1.0 m outside a window on the north wall of Norwood Hall, approximately midway along the length of the building. The height of the duct entrance from ground level was 9.1 m. The duct was aluminum, 7.0 m long × 5.0 cm diameter, with three 90° bends of 0.2 m radius. A centrifugal blower at the downstream end of the pipe gave sufficient suction to produce a flow of 300 L min^{-1}. The CCN counter sampled from this duct, at a point 1.0 m upstream of the blower. The Reynolds number in the duct was about 8×10^4, which is large enough so that the flow had to be turbulent. The losses of nuclei in this duct are estimated to be of order 1%, based on experiments performed by Hudson and Squires (1978) in a similar piping system.

An entire supersaturation spectra required about 45 min to complete, and was obtained once an hour, the first one of each day at about 0900 GMT, the last one at about 1700 GMT, so that generally, six spectra were taken per day. For the period 15 March–18 August 1978 this was done Monday through Friday. For the period 21 August–21 December 1981 this was done on Monday, Wednesday and Friday. A few exceptions to this rule occurred on holidays or during equipment breakdowns.

4. The data, average values and frequency distribution

The concentrations for the entire sampling period were averaged, with equal weight given to each hourly

[1] Royco Instruments, 141 Jefferson Ave., Menlo Park, CA 94025.

[2] Alofs, D. J., and M. B. Trueblood, 1981: UMR dual mode CCN counter. *Fourth Int. Workshop on Cloud Nuclei*, Reno, W. C. Kocmond, Ed., International Commission on Cloud Physics.

[3] Climet Instruments Company, 1320 W. Colton Avenue, Redlands, CA 92373.

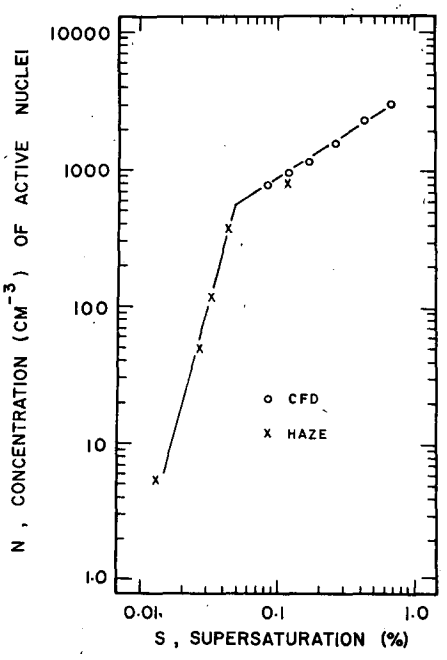

FIG. 1. Averaged CCN spectra.

data point. Fig. 1 shows these averages. The circles denote data taken in the CFD mode, whereas the symbol X denotes the haze mode. It can be seen from Fig. 1 that the haze mode concentration at $S = 0.122\%$ fell ~25% lower than the CFD concentration at the same supersaturation. The CFD value was judged to be more reliable and therefore was used in all of the analyses which are described below.

The most striking feature of Fig. 1 is that the averaged data can be seen to fall quite closely along two straight lines. Thus the averaged data are represented by the following two slope power law:

$$N = C_C S^{K_C}, \; S > S^*, \tag{2}$$

$$N = C_F S^{K_F}, \; S < S^*. \tag{3}$$

Following a suggestion by S. Twomey, we have assigned subscripts with mnemonic value in Eqs. (2) and (3). Thus subscript C suggests cloud condensation nuclei and subscript F suggests fog nuclei.

For the averaged data, the method of least squares was used to fit a line for the CFD mode data points, and another line for the haze mode data points, excluding the haze mode data at $S = 0.122\%$. The values of the parameters in Eqs. (2) and (3) thus computed are: $S^* = 0.0483\%$, $C_C = 3990$ cm^{-3}, $C_F = 6.616 \times 10^7$ cm^{-3}, $K_C = 0.643$, and $K_F = 3.85$. Note that the two slope power law probably does not apply in the region near S^*, especially because there is a gap in the data from $S = 0.044\%$ to $S = 0.083\%$. It is expected that the slope of the spectrum in reality changes gradually in this gap region.

It was decided to fit each of the 432 hourly spectra by a two slope power law. Many of the hourly spectra did not fit a power law as well as the averaged data. However, the power law approximation for $0.1\% < S < 1\%$ is well established by current practice. Also, the data in the fog nuclei regime was found to fit a power law at least as well as in the CCN regime. Moreover, it is a great convenience to represent the data by a two-slope power law.

The method of computing the power law for each hourly spectrum is now described using Table 1. The first column lists the supersaturations (S) at which data were always taken. The other columns show which values of S are assigned to Eq. (2) versus which are assigned to Eq. (3), for four different curve fits. For each curve fit, the method of least squares was used to calculate a residual, so that the best of the four curve fits was determined. The values from the best fit were then used to characterize that particular hourly CCN spectrum. The numbers in the parentheses in Table 1 show the percent of the cases each curve fit was the best. It can be seen that generally fit 2 was the best, as it also was for the averaged data.

Fig. 2 shows the frequency distributions for C_C, C_F, K_C, K_F and S^*. Note that Fig. 2 is a cumulative frequency distribution. For example, Fig. 2 shows that for 90% of the hourly spectra, $C_C < 10\,000$ cm^{-3}, i.e., for 10% of the spectra, $C_C > 10\,000$ cm^{-3}. This value of C_C will later be called the ninth decile value.

It also can be seen from Fig. 2 that the value of S^* is remarkably consistent. Indeed, 60% of the hourly spectra gave S^* in the narrow range 0.045–0.059%. A critical supersaturation of 0.05% corresponds to a 0.1 μm particle radius for pure ammonium sulfate particles (Twomey, 1977). Thus something rather abrupt and consistent is occurring near 0.1 μm radius.

5. The data—various correlations

In order to determine whether there were any discernible seasonal correlations in the data, monthly averages were computed. These averages are shown in Fig. 3 for the CFD mode, and in Fig. 4 for the haze mode. The values of S corresponding to each curve

TABLE 1. Values of S assigned to Eq. (2) and Eq. (3) for different curve fits of the hourly CCN spectra.

S (%)	Fit 1 (0)*	Fit 2 (77.5)	Fit 3 (22.2)	Fit 4 (0.3)
0.0133	↑	↑	↑	↑
0.0267	Eq. (3)	Eq. (3)	Eq. (3)	Eq. (3)
0.033	↓	↓	↓	↓
0.044	↑	↑	↑	↑
0.083				
0.122				
0.170	Eq. (2)	Eq. (2)	Eq. (2)	Eq. (2)
0.266				
0.436				
0.681	↓	↓	↓	↓

* Numbers in parentheses show the percentage of the cases each curve fit was the best.

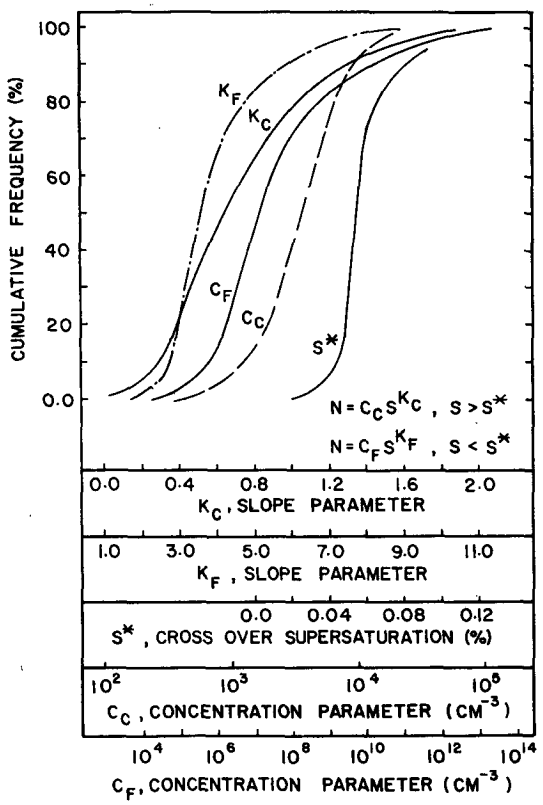

FIG. 2. Frequency spectra of each parameter in CCN spectra.

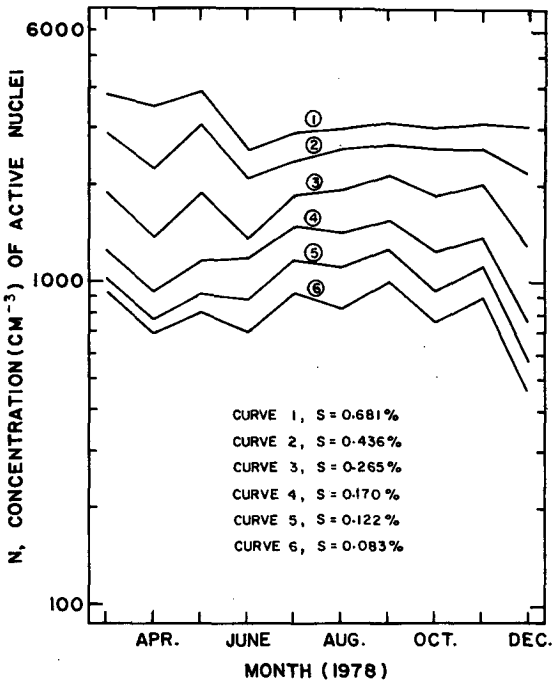

FIG. 3. Monthly average of concentrations for CFD mode.

are given on the figures. It can be seen from Figs. 3 and 4 that no seasonal correlation can be discerned. This result is in accord with the lack of a significant seasonal correlation reported by Twomey *et al.* (1978) for their 5-year program of monitoring CCN active at $S = 0.7\%$ in Australia.

Hourly surface wind measurements were available from the flight service weather station located at the airport near Vichy, Missouri, 23 km North of Rolla. An effort was made to correlate ths wind data to the measured CCN concentrations.

Fig. 5 shows the correlation with wind direction. The abscissa shows the direction the wind is blowing from, expressed in degrees measured clockwise from North. The ordinate shows the values of concentration of nuclei active at various values of S. The concentrations are averages over the entire data set. It can be seen that there is some correlation; however, the ratio of the maximum to minimum concentration is only about a factor of two on the average. This dependence on wind direction is considerably less than, for example, Twomey *et al.* (1978) report. Their ratio is about five to one. The maximum concentration is shown in Fig. 5 to occur when the wind is blowing approximately from the southeast. A possible explanation, is that, as mentioned previously, the world's largest lead mining operation is located 100 km southeast of Rolla and the refining operation associated with this district is known to produce sulfur dioxide.

Another possible correlation was investigated by comparing the average concentrations in the morning with those in the afternoon. The results are shown in Fig. 6. The ordinate shows the ratio of the average concentration in the morning, divided by the average concentration in the afternoon. The abscissa shows the supersaturation. Three curves are shown, corresponding to three different averaging time periods within the total data set. It can be seen that there is no strong correlation. In contrast, Twomey *et al.* (1978) and Jiusto and Lee (1975) reported that for supersaturation near 0.7%, the concentrations were ~10–40% higher in the afternoon than in the morning. Their diurnal trend was obtained by using only data for days when the wind was steady from one direction. This procedure allowed them to reach the interpretation that this trend was not due to diurnal changes in wind trajectory, but was instead likely caused by photochemical processes. Our data set had too few days of steady wind direction to extract a statistically meaningful correlation. Moreover, Fig. 6 suggests that no diurnal pattern exists in our data, hence there is no need to separate out the possibility of a diurnal pattern of wind direction.

6. Comparison with other CCN measurements

The measured value of C_C in the 1978 Rolla program is 3990 cm^{-3}, which is considerably higher than average values for continental air. For example,

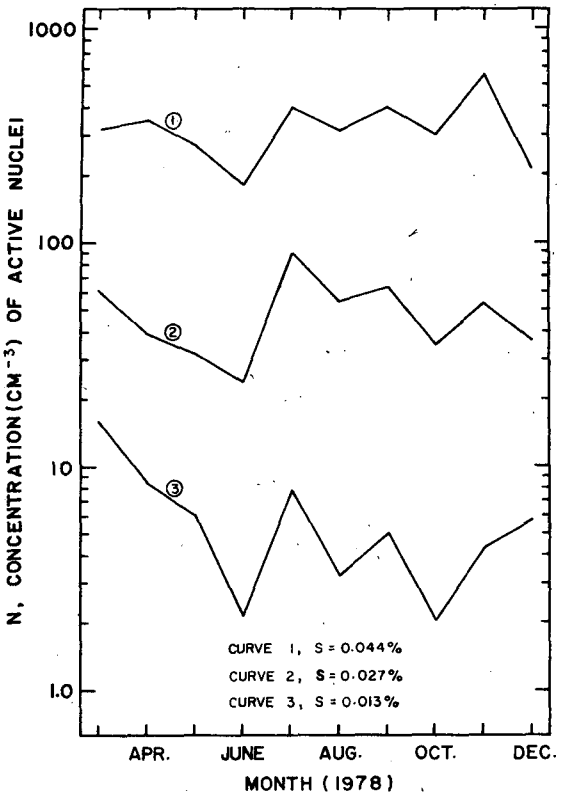

FIG. 4. Monthly average of concentrations for haze mode.

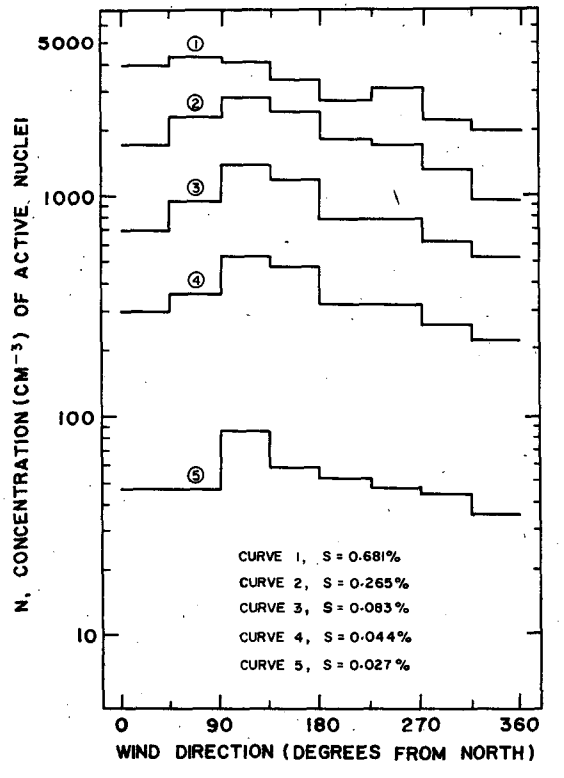

FIG. 5. Correlation between concentration and wind direction.

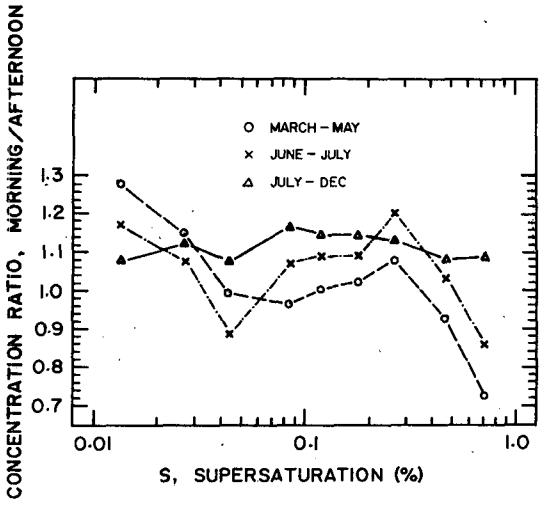

FIG. 6. Ratio of the average morning concentration to the average afternoon concentration.

Twomey and Wojciechowski (1969) report an average concentration of 400 cm^{-3} at $S = 1\%$ for long distance flights over Australia, Africa, and the United States. On the other hand, C_C values near 4000 cm^{-3} are not uncommon for urban areas. Consider for example, Kocmond's value (Jiusto and Kochmond, 1968) of 3300 cm^{-3} at Buffalo, New York; or the values of Hoppel *et al.* (1973) of 5400 cm^{-3} for measurements made 32 km south of Washington, D.C., and 9000 cm^{-3} for measurements closer to Washington, D.C. Fitzgerald and Spyers-Duran (1973) found average concentrations of 3890 cm^{-3} downwind of St. Louis, Missouri, and 2210 cm^{-3} upwind. A scatter diagram of their data shows values as high as 9000 cm^{-3}, which is close to the ninth decile value from the 1978 Rolla program. Likewise, Twomey (1963) reports maximum concentrations of 10 000 cm^{-3} over Australia.

The above examples are sufficient to show that the 1978 Rolla values are more typical of urban areas than continental averages. This is somewhat surprising because Rolla is a small town with very little industry. Local pollution sources are a possible explanation, but one would then expect stronger correlation with wind direction than the data exhibits (Fig. 5). Another possible explanation is that there are extensive forests in the vicinity; however, vegetation-produced CCN would exhibit a seasonal correlation, which the data does not show (Figs. 3 and 4).

Twomey (1959a) reports that near Sydney, Australia, the normal average concentration at $S = 1\%$ was 1000 cm^{-3}, but during drought conditions, the average concentration rose to 6400 cm^{-3}. Precipitation was about normal for Missouri during 1978, but perhaps other meteorological conditions were unusual. To investigate this possibility, additional

CCN measurements were made at Rolla for a 10-day period starting 14 July, 1981. These measurements were made at $S = 0.681\%$, continuously from 0800–1700 GMT, and yielded an average concentration of 2008 cm^{-3}, as compared to 3116 cm^{-3} at $S = 0.681\%$ during the 1978 program. Thus the 1981 measurements also show concentrations that are more typical of urban areas than continental averages. Three additional points concerning the 1981 measurements are now mentioned: first, simultaneous Aitken nucleus measurements averaged 14 900 cm^{-3}; second, the summer of 1981 was unusually wet; third, the same sample delivery pipe as in the 1978 program was used, and the CCN counter was unchanged except that the Climet OPC was used.

7. Survey of fog nuclei measurements

The only instrument suitable for making nuclei measurements in the range $0.01\% < S < 0.1\%$ is the haze chamber. Atmospheric measurements with this type of instrument are as yet quite few. The following discussion includes all those we are aware of.

Laktionov (1973)[4] made a series of measurements during the time frame 1966–72 using a CFD and the original haze chamber. Measurements from an aircraft at a height of 200 m over northeastern Russia showed very low S^*($S^* = 0.025\%$) with $K_C = 0.7$ and $K_F = 0.9$. Data taken near ground level at an unspecified location showed $S^* = 0.06\%$, $K_C = 0.8$, and $K_F = 2.7$. Measurements taken from a ship near the Atlantic tropical zone gave $S^* = 0.18\%$, $K_C = 1.0$, and $K_F = 2.7$. Thus Laktionov's measurements showed a considerable range of S^*, depending on geographic location.

Hoppel (1979) presented two CCN spectra obtained with the Naval Research Laboratory haze chamber and static diffusion chamber. Data taken 160 km off the New Jersey Coast showed $S^* = 0.2\%$, $K_C = 1.0$, and $K_F = 3$. Data taken in the Mediterranean gave $S^* = 0.03\%$, $K_F = 4$, and K_C varying smoothly from 1.0 at $S = 0.1\%$ to nearly zero at $S = 1\%$.

Hudson (1980) presented 12 CCN spectra obtained with the Desert Research Institute haze chamber and CFD. The measurements were made at several locations along the U.S. Pacific Coast. The values of K_F varied from 0.7 to 3.3, the average being 1.9. The CFD was used for only five of the 12 spectra, and for these five spectra, the average value of K_C was 0.89. Three of the spectra showed a rather abrupt change in slope at $S^* = 0.1\%$. Two showed only a gradual change of slope near 0.1% supersaturation. In one measurement taken at San Diego where the air mass was termed "polluted", both K_C and K_F had the same value (0.7).

8. Implications for aerosol size distribution or composition

Junge and McLaren (1971) investigated the relationship between CCN spectra and atmospheric aerosol size distributions. Four different model aerosol size distributions were used, models 1 and 2 being, respectively, the original Junge continental and maritime distributions, and models 3 and 4 being modifications of these having a dip in concentration at a radius r of 0.01 μm, in accord with more recent measurements. The volume fraction of water soluble material ϵ was assumed to not vary with particle size.

For $\epsilon \geq 0.1$, models 1 and 3 (continental) give CCN spectra with the following characteristics: $K_F \approx 2$, $K_C \approx 0$, and a very gradual transition in slope from K_F to K_C. Thus, our value of $K_F(3.85)$ is considerably higher than this prediction, and our change in slope occurs much more abruptly.

For $\epsilon = 1$, model 4 (maritime) gives CCN spectra with $K_F \approx 2.3$, $K_C \approx 0$, and a rather abrupt change in slope at $S \approx 0.05\%$. This suggests that one possible explanation for our abrupt change in slope would be an aerosol size distribution like model 4, having a strong peak at 0.1 μm radius. Unfortunately, aerosol size distributions were not measured in the 1978 program.

Another way to produce an abrupt change in slope near $S = 0.05\%$ is for ϵ to start decreasing for $r \leq 0.1$ μm. The following assumption turns out to be attractive: $\epsilon = 1$, $r \leq 0.1$ μm; $\epsilon = 0.1$ μm/r, $r \geq 0.1$ μm. This is because for models 1 and 3 (continental) it gives not only a rather abrupt change in slope at $S = 0.05\%$, but also a value of K_F in close agreement with our measured value. Moreover, Laktionov (1972b) and Mészáros (1968) found ϵ decreased in approximately this fashion.

9. Summary

Concentrations of CCN in the atmosphere were measured at Rolla, Missouri, near ground level. A total of 432 CCN spectra were measured in the supersaturation range from 0.013 to 0.681% during a 10-month period. On a logarithmic scale plot of active nuclei concentration (N) versus supersaturation (S), the data could generally be fitted well by two straight lines. The crossover supersaturation S^* where the two lines intersected was quite consistently near 0.05%. For 60% of the hourly spectra S^* was in the narrow range 0.045–0.059%.

The slope (K_F) of the straight line for $S < S^*$ had an average value of 3.85, whereas for $S > S^*$, the slope had an average value of 0.643. This large value

[4] Laktionov, A. G., 1973: Spectra of cloud nuclei in the supersaturation range 0.02–1%. *Proc. VIII Int. Conf. on Nucleation*, Leningrad, I. I. Gaivoronosky, Ed., International Union of Geodesy and Geophysics, 437–444.

of K_F suggests that for fogs and stratus clouds, nuclei concentrations may be much less important than in cumulus clouds.

The data did not show seasonal variation. Neither did they show the diurnal pattern which has been reported in two other CCN measurement programs.

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