

A Preliminary Numerical Study into the Effects of Coal Development on Cloud and Precipitation Processes in the Northern Great Plains

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ABSTRACT

Numerical simulations are conducted to determine the possible effects of air pollution from coal-fired power plants on cloud and precipitation processes in the northern Great Plains. This study can only be considered as preliminary in nature since a complete cloud simulation is not employed and the ice phase is not considered.

Natural and polluted particulate distributions are developed based on observations in the northern Great Plains and of coal-fired power plant effluent. Cloud droplet growth on these distributions is simulated in a condensation model. Results of this model indicate that the number concentration and dispersion coefficient (breadth) of the cloud droplet size distributions are increased by the addition of pollutant particles, especially if these are more hygroscopic than the background nuclei.

Coalescence calculations using the results of the condensation studies as input are also reported. These results indicate that the rate of production of large drops, while being slowed by an increase in the number concentration, is hastened by an increase in the dispersion coefficient. These two effects nearly cancel each other out so that the time required for precipitation development is very nearly the same for cloud droplet distributions initialized on background and polluted particulate distributions. If, however, both distributions have the same dispersion, the polluted case requires a considerably longer time to develop precipitation.

1. Introduction

Recent concern about energy sources has prompted plans for the large-scale development of western coal reserves. Attendant to this development, construction and operation in the northern Great Plains area of mine-mouth, coal-fired power generating plants with energy outputs in the thousand megawatt range (Atmospheric Aspects Work Group, 1974) are likely. Since the northern Great Plains region has a semi-arid climate, and most agriculture is "moisture limited," the possible effects of pollution released from these facilities on cloud and precipitation processes are of considerable interest. Numerical simulations using cloud and condensation models represent one means of investigating these effects.

In an earlier numerical study of effects of pollution on condensational growth, Chen and Davis (1974) found that large concentrations of small nuclei tend to reduce the droplet mean radius and the concentration of large drops. They concluded that this would tend to reduce the likelihood of precipitation by way of the coalescence process, but they did not include a coalescence calculation. A cloud model simulation,

including both condensation and coalescence, of the effects of condensation nuclei from a paper mill upon warm clouds in Washington state was made by Hindman and Hobbs (1974). They found, in agreement with observations, that the addition of large, highly hygroscopic nuclei resulted in a drop spectrum which was broader than that resulting from the ambient nucleus distribution. More recent work (Hindman *et al.*, 1977a) has also shown that increases in the concentration of small CCN with the concentration of large CCN held constant results in a narrowing of the droplet distribution. They concluded that high concentrations of small CCN produce narrow droplet spectra regardless of the concentration of large and giant CCN. In a study of CCN and cloud droplet size distributions in and around paper mills, Hindman *et al.* (1977b) reported on model calculations which roughly duplicated the observed changes in the cloud droplet spectra, particularly the increase in concentration at the large droplet tail of the spectra, resulting from the large and giant CCN emitted by the paper mills. These studies indicate that differences in the nucleus size distribution and solubility may lead to differences in cloud droplet spectra and, possibly, formation of precipitation.

This paper is a study of the possible effects of particulate pollution on coalescence rain development in clouds of the northern Great Plains. Particle dis-

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tributions representative of natural background levels in the northern Great Plains and of pollution appropriate to that produced by coal-fired power plants are developed. Various mixtures of these distributions, representing different levels of pollution, are used as input to a condensation model. The effects of pollution on the resulting cloud droplet spectra are discussed. The cloud droplet spectra are characterized by their dispersion coefficient and number concentration to define an initial distribution for coalescence calculations. Based on the results of these calculations, comments on the effects of pollution on precipitation formation by the coalescence process are made. Actual precipitation production, in addition to usually involving the ice phase for clouds in the northern Great Plains, is the result of poorly understood interactions between the microphysics and dynamics well beyond the scope of this study.

2. Particle distributions and chemistry

Aerosol particle distributions for input to the condensation model were obtained by adding pollutant particles to a background distribution. To obtain realistic, useful distributions, information on both the size distribution and chemical composition for natural and pollutant particles was needed. However, for the situation under consideration, data were scarce and uncertain, resulting in some arbitrariness in the distributions. Also, as a matter of convenience, we shall frequently refer to power plant effluent as pollution or pollutant particles throughout the text.

a. Background distribution

Measurements of natural aerosols in the 0.15–5.0 μm radius interval were made below cloud base in western Nebraska and northeastern Colorado by Blifford and Ringer (1969) and Hidy *et al.* (1970). Total particulate concentrations in this size range were quite low and ranged from 1–10 cm^{-3} .

Since measurements of small condensation nuclei were not available for this area, cloud droplet concentration measurements were used to fix the small end of the particle distribution. Marwitz and Auer (1968) measured updraft velocities and activated cloud nucleus concentrations for a variety of clouds in northeastern Colorado and South Dakota. Their results indicate that updraft velocities of 3 m s^{-1} and activated nucleus concentrations on the order of 500 cm^{-3} can be considered average conditions. The activated nucleus concentrations they obtained agree with other measurements of cloud droplet measurements in the same area. Miller *et al.* (1967) found 400–600 cm^{-3} and Takeuchi (1971) and Schroeder (1973) found 200–800 cm^{-3} .

The background particle size distribution used in this study was designed to fall in the vicinity of the

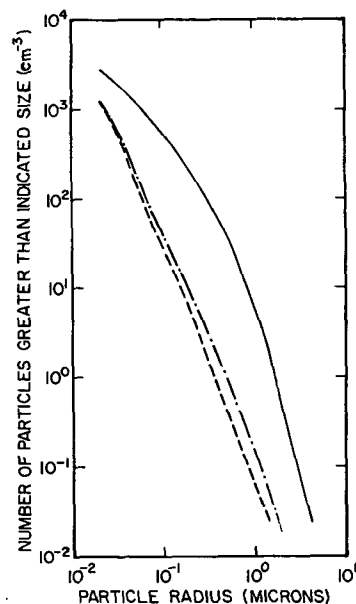


FIG. 1. Cumulative particle size distribution used as input to the condensation model. The dashed line is the background distribution, the dot-dash line the total distribution (background plus pollution) 12 km downwind of the pollution source, and the solid line the total distribution 1 km downwind of the source.

measurements of Blifford and Ringer (1969) and Hidy *et al.* (1970) for the larger particles and to give 400–600 activated nuclei with a 3 m s^{-1} updraft. Although there is evidence that particulate distributions actually have a complex form (Graedel and Franey, 1974; Shettle, 1975; Lee and Juisto, 1974; Juisto and Lee, 1975), the lack of detailed data throughout the entire size range of interest to condensation studies has led most investigators to use an average smooth curve distribution. This method was used in this study also. The resulting distribution is shown in Fig. 1 (dashed line). This distribution is similar to the clean continental type distributions used by Neiburger and Chien (1960) and Chen and Davis (1974). The largest nucleus considered in our study has a radius of 9 μm . The studies of Takahashi (1976) and Hindman *et al.* (1977a) indicate that nuclei beyond about 10 μm radius are unimportant with regard to activated number concentration and dispersion of the resulting cloud droplet distribution. Takahashi has also suggested that giant nuclei are not responsible for the formation of raindrops but are important for chemical balance.

The assumed partial solubility of the background nuclei in this study is based on the following studies. Hidy *et al.* (1970) made a microscopic study of large and giant aerosol particles and reported that as many as 80% of the particles contained ammonium sulfate, and no more than 15% contained sea salt. Rosinski and Nagamoto (1972) sampling in Colorado similarly found that large percentages of the particles in the

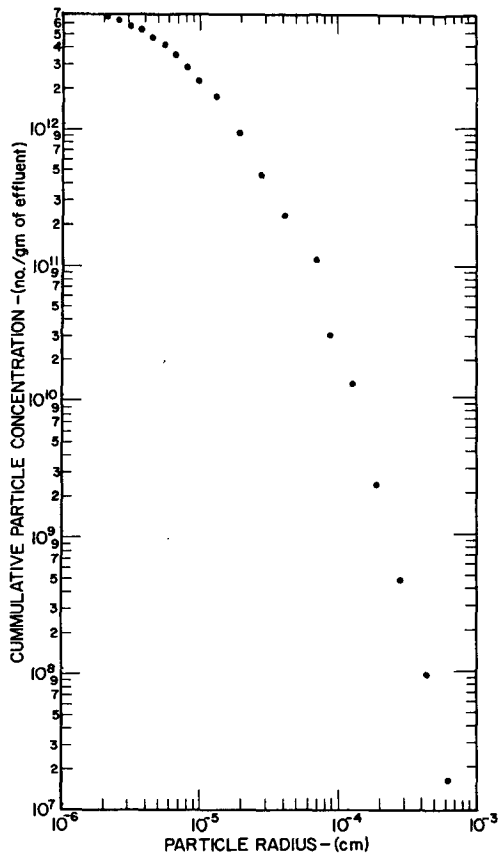


FIG. 2. Cumulative particle distribution of pollution per gram of effluent released by a typical coal-fired generating plant.

1.5–9.0 μm diameter range were hygroscopic, but only small percentages contained chlorides. Rinehart (1971) found that over an isolated New Mexico site, most giant particles were partly soluble and that the soluble content of large and giant particles was usually less than 10% of the total mass. In aerosols sampled over Hungary, Meszaros (1968) reported that the ratio of soluble to insoluble substances was on the order of 20% for Aitken sized particles and decreased to about 5% or less for larger particles, and suggested that the major water soluble component was ammonium sulfate. Laktionov (1972) obtained similar results from samples taken over Russia. Thus, for this study each particle was assumed to be partly soluble: 20% if less than 0.1 μm radius, and decreasing to 5% for particles greater than 1 μm radius. The soluble component was assumed to be ammonium sulfate.

b. Pollutant distributions

As was the case for the background distribution, information used to develop the pollutant distributions had to be gleaned from a variety of sources. McCain *et al.* (1975) determined detailed particle size distributions for particles of less than 5 μm radius emitted by coal-fired power plants, both with and

without high efficiency electrostatic precipitators. The overall mass efficiency of the precipitator in the McCain study was 99.6%. The particle size distribution was extended to larger sizes using the data of Shannon *et al.* (1970). The effluent particle size distribution (post-precipitator) for a coal-fired power plant equipped with a high efficiency electrostatic precipitator as in the McCain study is displayed in Fig. 2. Here the distribution is presented as the cumulative concentration per gram of effluent released to the atmosphere.

The chemical composition of the pollutant distributions must also be specified. The ash content of Wyoming coal ranges between 10 and 15% by weight, while the sulfur content ranges between 0.4 and 0.8%. The fly ash is composed mainly of the oxides of silicon, aluminum, iron and calcium with lesser amounts of the oxides of titanium, magnesium, potassium, sodium, lithium and phosphorus plus soluble sulfates (Cook, 1975; and Bickelhaupt, 1975). Emission standards [New Source Performance Standards (NSPS)] imposed on new power plants of greater than 25 MW capacity limit particulate emissions to 0.1 pounds per million BTU ($\sim 4 \times 10^{-7}$ gm J^{-1}). Limitations for SO_2 and NO_x emissions, on the other hand, are 1.2 and 0.7 pounds per million BTU, respectively. Specific controls on SO_2 emissions may remove as much as 90% of the SO_2 from the effluent. Control technology for NO_x is much less advanced, however, and usually relies on low excess air to keep the combustion temperature and, hence, NO_x emissions down (Atmospheric Aspects Work Group, 1974). The fly ash will adsorb some of the SO_2 released, so some will be removed by the electrostatic precipitator. This collection mechanism plus the low sulfur content of northern Great Plains coal usually result in SO_2 emissions being well within the NSPS limit, even without specific controls for SO_2 .

As the preceding discussion indicates, the determination of the chemical composition of power plant effluent is a complex problem. It would be most desirable to know the chemical composition and solubility of power plant effluent as a function of size. Due to the lack of measurements of these characteristics, the assignments we have made for both chemical composition and solubility are quite arbitrary. It is known that coal-fired power plants produce large amounts of sulfuric acid. For this study we assume that the soluble component of the pollution is sulfuric acid for particles $< 1 \mu\text{m}$ radius. For larger particles, sodium sulfate is assumed to be the soluble component. The pollutant aerosols below 0.1 μm radius are assumed to be 40% soluble, decreasing to 5% soluble for particles over 1 μm radius. These choices account for a considerable portion of the SO_2 released.

For this study, we assume that all of the power plant effluent has a hygroscopic component, being the

result of interactions between the fly ash and SO_2 emitted. Gas-to-particle conversion processes and/or catalytic oxidation of the SO_2 are thus tacitly assumed. The role of these processes in the formation of sulfates has been investigated by numerous workers, usually studying other types of pollution. Eagen *et al.* (1974) noted increases in CCN concentrations downwind of Kraft paper mills and attributed the increases to gas-to-particle conversions. Freiberg (1976) suggested that relative humidity and temperature strongly influence the gas-to-particle conversion process. Novakov *et al.* (1974) studied the catalytic oxidation of SO_2 on soot particles. They concluded that this process played a major role in sulfate formation in urban atmospheres, but also noted that it is especially likely in or near combustion sources. Pueschel (1976) reported on airborne measurements taken in the plume of the Four Corners power plant. He concluded that a stable form of sulfur accumulated on the surface of submicron fly ash particles by adsorption and oxidation before a gas-to-particle conversion process could take place. Whatever their form, these processes are likely to be critically important with regard to pollutant particle chemistry. Although these processes are implicit in the degree to which sulfur forms are assumed in the pollutant chemistry specified above, such reactions could also result in forms of NO_x being incorporated with the pollution particles. Due to the complexity of the air pollution chemistry involved, however, we will not address this problem in detail but, instead, will incorporate into the model the simpler chemistry noted above.

The effluent will be carried downwind of the power plant and will eventually be attenuated to background levels. Downwind concentrations were calculated using a simple Gaussian plume model (Turner, 1970). For this study, we have assumed a 1000 MW power plant and a Pasquill C atmospheric stability category. (This stability category corresponds to conditions of moderate to strong incoming solar radiation with surface wind speeds of 5 m s^{-1} .) The total particle size distributions (background plus pollution) for distances of 1 and 12 km downwind of the power plant are shown in Fig. 1. The required output, uncontrolled, to produce these distributions is slightly more than 10 kg s^{-1} of particulates. It is worth noting here that the emissions for this hypothetical power plant are about 35% of that allowed by the NSPS issued by the Environmental Protection Agency. This is a direct result of assuming the presence of high-efficiency electrostatic precipitators. Actually, the low sulfur coal common to the northern Great Plains region creates charging problems in electrostatic precipitators if run at full current. Efficiency is reduced if the current is reduced to overcome these difficulties. Therefore, it seems that our distributions may be overly optimistic in terms of attainable concentrations.

Fortunately, large changes in source strength are required to make significant changes in downwind concentrations for the stability category we have assumed. The distance downwind of the source for a given concentration to exist is a function of source strength, wind and stability, with wind and stability being very important. Thus, changes in these variables merely represent a shift upwind or downwind for finding a given concentration. For a more stable air mass than we have assumed here, the concentration comparable to our 1 km polluted case may exist 3–4 km or more downwind of the source.

The nucleus distributions developed above as being appropriate for a mixture of power plant effluent with the background nuclei at various distances downwind of the source will hereafter be referred to as polluted distributions. Recall from our earlier discussion that, due to the lack of detailed measurements on the characteristics of power plant effluent, certain assumptions were made with regard to the size distribution and chemical properties of the effluent. Other nucleus distributions, referred to as Type II and Type III distributions below, were therefore developed to investigate the effects of the shape of the size distribution and chemical composition of the power plant effluent on cloud droplet formation.

The solubility of the background nuclei is less than that of the power plant pollutant particles. Therefore, to study the effects of pollutant particle solubility, the condensation model was also run using nucleus populations (Type II) having the same size distributions as those of the polluted atmosphere, but with the pollutant particles having the same chemical composition and solubility as the background nuclei.

The shape of the size distribution of the pollutant particles from the power plant is not similar to that of the size distribution of the background particles. A third type of distribution (Type III) was developed assuming that the pollutant particles had both the same shape of size distribution and the same chemical composition as the background nuclei but in concentrations comparable to the polluted cases. Thus, the effect of the shape of the pollutant particle size distribution on cloud droplet formation is illustrated by comparison of cloud droplet spectra initialized on these Type III nuclei distributions with droplet spectra grown on Type II distributions. Therefore, the condensation model was run using all three types (Polluted, Type II, Type III) of distributions at various "pollution levels" for input.

3. Condensation model

The condensation model is a modification of one developed by Chen (1971). The model permits interacting cloud microphysics and dynamics in a one-dimensional, single-parcel, Lagrangian framework with entrainment. Aspects of this model were discussed in

two notes (Chen, 1974a,b). The model has been modified to consider a spectrum of nuclei of different chemical compositions and to conserve computer time. Droplet growth in this model is controlled by the growth equation

$$\frac{dr}{dt} = \frac{D}{\rho_s r} \left\{ \rho_\infty - \rho_{\text{sat}} \left[\exp\left(\frac{2\sigma}{\rho_s R_s T r} - \frac{m_w}{1000} M \nu \phi + \frac{\rho_s r (dr/dt) L_v}{R_s T^2 K} \right) \right] \right\}, \quad (1)$$

where r is the droplet radius, D the diffusion coefficient of water vapor, ρ_∞ the ambient vapor density, ρ_{sat} the saturation vapor density over a flat water surface and ρ_s the solution density. Additionally in (1), σ is the surface tension of the solution, R_s the gas constant for water vapor, T the air temperature, L_v the latent heat of vaporization and K the thermal conductivity. Symbols representing vapor pressure depression due to the solution effect are m_w , the molecular weight of water, ν , the number of dissociated ion species of the solute, and M and ϕ , the molality and osmotic coefficient, respectively, of the solution.

For the calculation of the diffusion and heat coefficients, the accommodation and condensation coefficients must be specified. The proper values for these coefficients have been the subject of a long and continuing debate. In this study the value of 0.7 was chosen for the accommodation coefficient consistent with the work of Fitzgerald (1974). The condensation coefficient was set at 0.033 in agreement with recent experimental results (Sinnarwalla *et al.*, 1975; Chodes *et al.*, 1974). Otherwise, Chen's (1974b) formulation has been retained.

Chen's original model used refined numerical integration techniques of the predictor-corrector type which required very short time steps and considerable amounts of computer time. This scheme has been discarded in favor of a simple Eulerian solution with the restriction that the smallest droplets, which tend to grow or shrink unrealistically fast numerically, are put near their equilibrium values. This method was introduced by Mordy (1959) and later refined by Berry and Reinhardt (1973), whose treatment is followed here. This has enabled the use of vastly larger time steps and has resulted in a considerable savings in computer time.

The equilibrium technique assumes that the growth is always in the direction of the equilibrium (Köhler) curve. The increment of growth, Δr , resulting from (1) is compared to the growth required to reach the equilibrium radius (r_{eq}). If $\Delta r > 0.2(r_{\text{eq}} - r)$, then a modified smaller Δr is calculated according to

$$\Delta r|_{\text{mod}} = (r_{\text{eq}} - r) \left[1 - \exp\left(\frac{\Delta r}{r_{\text{eq}} - r} \right) \right]. \quad (2)$$

This formulation allows the rate to decay exponentially as the radius approaches the equilibrium value. The equilibrium radius used above is computed by Newton's method for all droplets having a radius $< 0.4 \mu\text{m}$ and which are smaller than the critical radius. Droplets larger than the critical radius are generally in a regime of stable growth and do not require this stabilizing technique.

In this study, only soluble or partly soluble particles are treated. Totally insoluble particles are not considered; however, the results of studies by Mahata and Alofs (1975) and Junge and McLaren (1971) show that it is unlikely that such particles are important in atmospheric condensation. The soluble portion may be varied between categories and may be of mixed composition. The surface tension, droplet density and the vapor pressure reduction over a solution are all considered as functions of droplet radius and composition.

The solution effect on vapor pressure over a droplet is treated as described by Chen (1974a); this may be written as

$$\frac{e_s}{e_\infty} = \exp\left[-\left(\frac{m_w}{1000} \right) \left(\sum \nu_i M_i \phi_i \right) \right], \quad (3)$$

where e_s refers to the vapor pressure over the solution with e_∞ being the vapor pressure over a pure water surface. The other symbols are as defined earlier, with the summation over i indicating the total contribution of all soluble components. Since the $m_w/1000$ factor is a constant, it may be taken inside the summation and we may define

$$Z_i = \left(\frac{m_w}{1000} \right) \nu_i M_i \phi_i. \quad (4)$$

This quantity may then be found as a function of molality for any compound for which osmotic coefficient data are available. A simple summation permits treatment of mixed solutions. For this study, a polynomial regression for Z_i is used, employing up to six terms. Robinson and Stokes (1970) is an excellent source of osmotic coefficient data for many electrolytes, although references for others can be found in Pitzer and Mayorga (1973).

The surface tension, in addition to being a function of temperature and droplet radius, is also modified by the presence of soluble components, being dependent on the molality of the solution. Lack of data on the effect of composition on surface tension is, as Low (1969) found, a major problem. The lack of a simple formulation for combining effects in the case of a mixed solution also represents a major limiting factor. One saving point here is that the condensation model results are quite insensitive to the effect of composition on surface tension.

4. Condensation studies

The model computations began at a pressure of 885 mb, a temperature of 20°C and a relative humidity of 99%. All nuclei were assumed to be in equilibrium at this point. An initial updraft velocity of 3 m s⁻¹ was used, and the model was run to 40 s of simulated real time. This represents a rise to approximately 100 m above cloud base (100% relative humidity). For computations, the background and pollutant particle contributions were both discretized into 97 initial size classes which are then handled in a Lagrangian sense. The maximum supersaturation and the number of nuclei which are activated are only accurate within the limits marked off by the smallest size class of nuclei to be activated and the largest nucleus to remain unactivated. The dispersion coefficient³ of the cloud droplet size distribution is critically affected by this accuracy. The 97 size classes used contained no more than 19 particles in any single size class for the background distribution. For the cases discussed here, the use of 97 classes is sufficient to result in number concentration being resolved to within 3% with estimates of the dispersion coefficient likely to be off by as much as 5% and mean radius <2%. The scatter in the data in Fig. 4 (discussed later) can be considered to be an indication of the effect of this problem.

The results from the condensation model after 100 m rise above cloud base (100% relative humidity) are displayed in Fig. 3. The calculated droplet spectrum initialized on the background distribution is shown in Fig. 3a, while the resulting droplet spectra at different distances downwind of the pollution source are shown in Figs. 3b-3d. Fig. 3b presents the cloud droplet distribution 12 km downwind; Fig. 3c, the distribution 3.5 km downwind; and Fig. 3d, the distribution 1 km downwind. For the polluted cases (Figs. 3b-3d), the contributions from the background (dash-dotted lines) and pollution (dashed lines) are shown, as well as the combined or total distribution (solid lines). We note in Fig. 3b that the pollution is providing only a minor contribution (about 5% of the total number of activated nuclei, mainly for droplets between 6 and 10 μm diameter) to the resulting distribution. In Fig. 3c, the background and pollution provide nearly equal contributions (45% pollution, 55% background), with the pollution providing the nuclei for droplets in the larger sizes, while the background supplies the bulk of the nuclei for the smaller droplets. For the situation 1 km downwind

TABLE 1. Summary of condensation model results.

Parameter	Back-ground	Polluted cases		
		12 km down-wind	3.5 km down-wind	1 km down-wind
Number concentration (cm ⁻³)	524.5	531.1	610.5	1268
Dispersion coefficient	0.127	0.135	0.163	0.237
Mean diameter (μm)	7.38	7.32	6.94	5.34
Smallest activated nucleus (dry radius, μm)				
Background	0.0308	0.0311	0.0361	0.0527
Pollution	—	0.0235	0.0275	0.0410

of the pollution source (Fig. 3d), the pollution provides the vast majority (~90%) of the activated nuclei.

Table 1 summarizes other aspects of the results from the condensation model for the four cases in Fig. 3. The number concentration and dispersion coefficient of the cloud droplet spectra are seen to increase as the level of pollution increases, while the mean diameter decreases. The smallest activated nucleus from the background and that from the pollution are also listed. As the number of available nuclei increases, the size of the smallest activated nucleus also increases. Due to the higher degree of hygroscopicity of the pollution particles, the smallest activated nucleus from the pollution is nearly 25% smaller (in radius) than the smallest nucleus activated from the background. In all cases, the condensation model indicates a narrow distribution with small values (<0.25) for the dispersion coefficient.

Warner (1969a) reported on observations of cloud droplet spectra taken over an eight-year period. He was unable to discern any relationship between droplet number concentration and dispersion coefficient and concluded that these features of the cloud droplet size distribution were independent, especially near cloud base. He also reported on results of the integration of the condensation equation (Warner, 1969b). He found the theory to be adequate in describing most observed features of the droplet distribution with the exception of the dispersion, for which the theory consistently predicted lower values than were observed. This discrepancy between measurements and theory has received much attention with regard to both condensational growth theory and an examination of physical effects which may result in a broader cloud droplet size distribution. An excellent review of work to explain and understand this discrepancy is given in Rogers (1976).

We shall now attempt to determine the influence of the shape and solubility of the assumed polluted distributions used above. Recall from the discussion of the polluted distributions that two other versions of the distributions are to be investigated. The Type II

³ The dispersion coefficient (=σ/d, where σ is the standard deviation and d the mean diameter of the distribution) is a measure of the breadth of the distribution. This quantity will frequently be referred to as dispersion, but in all cases will refer to the statistical spread of the distribution rather than in the sense of diffusion of material in the atmosphere.

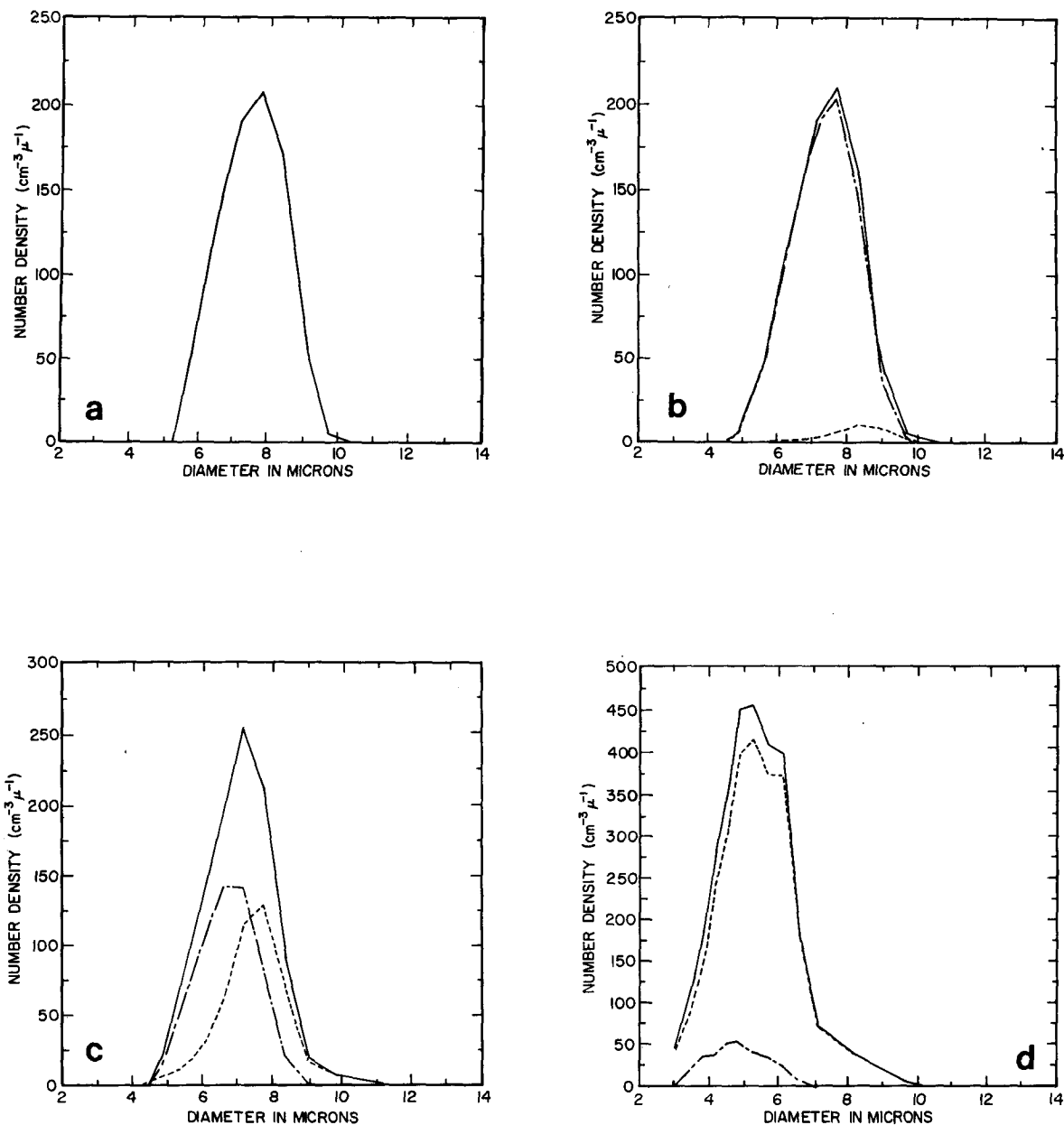


FIG. 3. Cloud droplet distributions predicted by the condensation model after 100 meters rise above cloud base. Particle size distributions used as input are: (a) background, (b) background plus pollution 12 km downwind of source, (c) background plus pollution 3.5 km downwind, and (d) background plus pollution 1 km downwind. For the polluted distributions (b)–(d), the dashed lines represent the contribution from the pollution, the dash-dotted lines represent that from the background, and the solid lines represent the sum of these two contributions. See Table 1 for further information on these distributions.

distribution assumes that the particles have the same size distribution used for the fully developed polluted distribution, but the same chemical composition as the background. Type III distributions assume the same shape and chemical composition as the background distribution, although the concentrations are similar to the polluted distributions. A comparison of the results for model computations of all three types of distributions gave the following results. In all

cases, increasing the particle number concentration resulted in an increase of the resulting cloud droplet number concentration and a decrease in droplet mean radius. The decrease in mean radius with increasing number concentration was not affected by pollutant particle size distribution or solubility.

From a graph of droplet dispersion coefficient versus droplet number concentration (Fig. 4) it can be seen that the effect of pollution on droplet dispersion is

dependent on both the shape of the particle size distribution and its chemical composition. In all cases, the droplet dispersion tends to increase with increasing number concentration. The greatest increase is exhibited with the addition of larger and more hygroscopic pollutant particles. The addition of particles which were larger but with the same chemical composition as the background (Type II distributions) gave a smaller increase in droplet dispersion with increasing number concentration. The smallest increase was obtained for simple increases in number concentration without changing the shape of the particle size distribution or solubility (Type III distributions). Thus, the model predicts that the addition of pollutant particles to a clean continental background distribution tends to increase droplet dispersion, at least for the distributions we have assumed. The largest increases are obtained if the added particles are both larger and more hygroscopic than the background (efficient cloud condensation nuclei). One could also infer that if the nuclei resulting from the pollution were less hygroscopic than the background nuclei or were effective over only a limited size range, a decrease in dispersion might result. The effect of particle solubility is similar in many respects to that obtained by Fitzgerald (1974).

An explanation for these results is offered. The increase in droplet dispersion for the polluted distributions can come about as the result of the increased number of larger particles. The number of larger droplets will be increased while decreasing the size of the smaller droplets as they compete for the available water vapor. This effect will be enhanced if the added larger particles are also more hygroscopic. This can be seen in Fig. 3 by the shift to smaller sizes by the small end of the droplet distribution, while the largest size remains fairly well fixed as the degree of pollution is increased. The tendency of droplets resulting from the pollution to be in the larger sizes is clearly shown in Fig. 3b and 3c. Although Fig. 3d indicates fewer large droplets than the other cases, the nuclei resulting from the power plant effluent are responsible for most droplets, especially those greater than $7 \mu\text{m}$ radius. A similar explanation was proposed by Hobbs *et al.* (1970) to explain the increased precipitation downwind of paper mills in Washington state. Observations by Eagen *et al.* (1974) and a numerical simulation by Hindman and Hobbs (1974) provided corroborating evidence.

5. Coalescence calculations

Twomey (1966), Berry (1968) and Berry and Reinhardt (1974) investigated the effects of cloud number concentration and the dispersion coefficient of the cloud droplet distribution on coalescence rates. Number concentration was found to have the greater

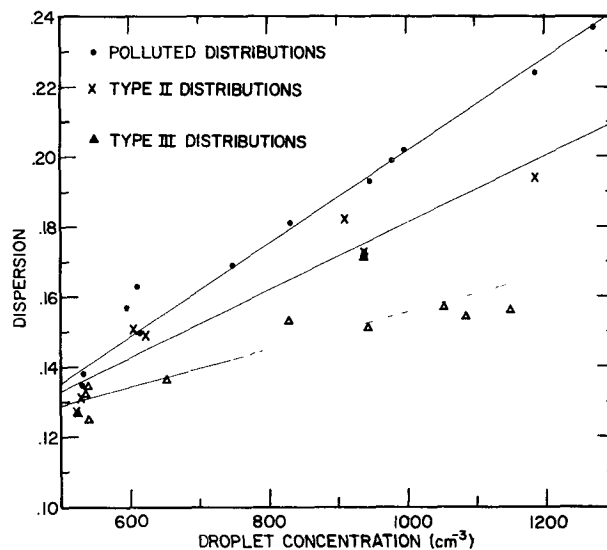


FIG. 4. Dispersion coefficient versus droplet number concentration showing effect of shape and composition of the pollutant distribution. Polluted distributions are those as described in Section 2b. Type II distributions assume the same shape as the polluted distributions but the composition of the background. Type III distributions have the same shape and composition as the background but in greater numbers.

influence on the rate of production of large drops with high number concentrations resulting in slower rates. A narrow dispersion was also found to slow the rates, but dispersion was found to be less important than the number concentration. Because the condensation studies have indicated increases in number concentration and a larger dispersion in the presence of pollution, it becomes difficult to make a judgment on the influence pollution will have on the precipitation process. Therefore, we have performed coalescence calculations to help determine the influence of power plant effluent on precipitation development. Leighton and Rogers (1974) have shown that a combination of condensation and coalescence leads to much faster development of precipitation than the coalescence process acting alone. It thus appears that any future work should allow for these two processes operating in concert.

For these coalescence calculations we have employed the formulation used by Farley and Chen (1975), which includes drop breakup. The droplet number concentration and dispersion determined from the condensation studies are used in specifying the gamma distribution which describes the initial droplet spectra (Borovikov and Khrgian, 1961; Clark, 1974). The time rate of change of the size distribution due to stochastic coalescence is evaluated by the method introduced by Kovetz and Olund (1969) and computationally improved by Nelson (Silverman and

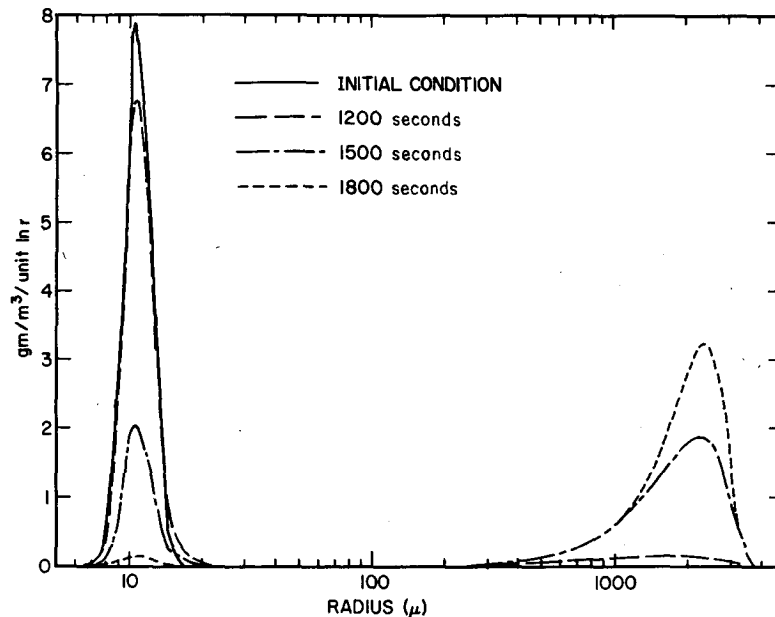


FIG. 5. Computed droplet growth due to coalescence and drop breakup (disintegration type) for a cloud droplet distribution resulting from the background particle distribution. The initial distribution has a number concentration of 524.5 cm^{-3} and a dispersion coefficient of 0.127.

Glass, 1973). Large drops are allowed to disintegrate according to the treatment of Srivastava (1971).⁴

Three droplet spectra are considered—those resulting from the background and 1 km polluted cases, and a modified background case using the same dispersion as the polluted case. The initial cloud droplet spectra for these three cases differ quite markedly with the intent purpose of trying to assess changes in the coalescence process caused by a very high pollution level. The results of these calculations are plotted in terms of the mass density function introduced by Berry (1967). A liquid water content of 2.5 g m^{-3} was used for all cases. The narrow initial distribution of the background case (Fig. 5) is clearly evident when compared to the polluted (Fig. 6) and the modified background (Fig. 7) cases. The larger mean diameter of the initial distribution of the background and modified background is also quite evident. Additional curves on each figure show the distributions at various stages of simulated development. The large drop end of the curves exhibits a steep cutoff as a result of drop breakup. A fourth case assuming the number concentration of the polluted distribution and the dispersion resulting from the background was also used. The growth rates were extremely slow in this case and are not displayed.

⁴Subsequent to the writing of the thesis, work by List and Gillespie (1976) indicates that a breakup scheme based on collision-induced breakup [similar to that used by Young (1975)] is more appropriate. Use of a breakup scheme of this type is not expected to change the conclusions of this paper.

The curves describing the distributions at various stages permit a comparison of growth rates. The droplet population for the background case has grown almost as much in 1500 s as has that of the polluted case in 1800 s. The modified case at 1200 s has greatly surpassed this stage of development and has almost reached the same level of growth as the background case at 1800 s. Thus growth for this case is significantly greater. The slow rate of growth of the background case relative to the growth rate of the modified background case is in agreement with the results of previous studies showing that a narrower distribution slows the coalescence rate. Comparison of the polluted case to the modified background case shows that the larger number concentration and smaller mean diameter of the polluted case significantly slow the coalescence rate. However, comparison to the background case shows that the larger dispersion of the polluted case largely overcomes the effects of larger number concentration and smaller mean diameter. Thus, the polluted case and the background case have similar coalescence rates. These results show that the effect of pollution on the dispersion coefficient of the cloud droplet distribution can be important in determining the effect of pollution on coalescence rates and may largely offset the negative effect of an increase in number concentration. If, however, as the observations of Warner (1969a) indicate, the dispersion is fairly constant regardless of number concentration, then pollution can be expected to retard the development of warm rain precipitation.

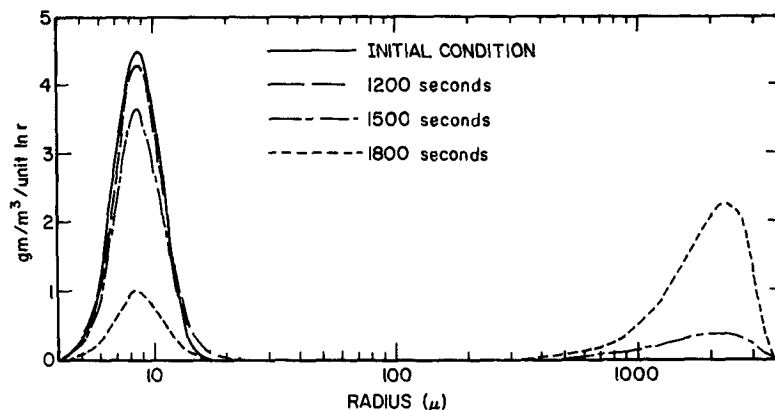


FIG. 6. As in Fig. 5 but for the cloud droplet distribution resulting from the 1 km polluted distribution. The initial distribution has a number concentration of 1268 cm^{-3} and a dispersion coefficient of 0.237.

6. Summary

Particle distributions representing a clean continental type background and polluted conditions resulting from power plant effluent were developed. These were used in a condensation model to study the effects of this form of pollution on cloud droplet formation. For the conditions simulated, an increase in nucleus concentrations resulted in an increase in cloud droplet concentration and a decrease in droplet mean diameter. The shape of the nucleus size distribution and chemical composition had little apparent effect on number concentration or mean diameter. The dispersion coefficient of the cloud droplet distribution, however, was found to be dependent on both the shape and chemical composition of the pollutant particle distribution. The

largest dispersions were obtained for pollutant particles which were both larger and more soluble than the background. Coalescence calculations showed that a larger droplet number concentration and smaller mean diameter tend to slow the coalescence rate, but a larger dispersion coefficient of the droplet distribution tends to hasten coalescence. Thus the effect of pollution from coal-fired power plants on the dispersion of the cloud droplet distribution is important in determining its effect on coalescence.

For the modeled power plant effluent, these effects tended to balance and only a small decrease in the coalescence rate was observed for the 1 km polluted case compared to that resulting from the background case. As warm clouds produce rain primarily by

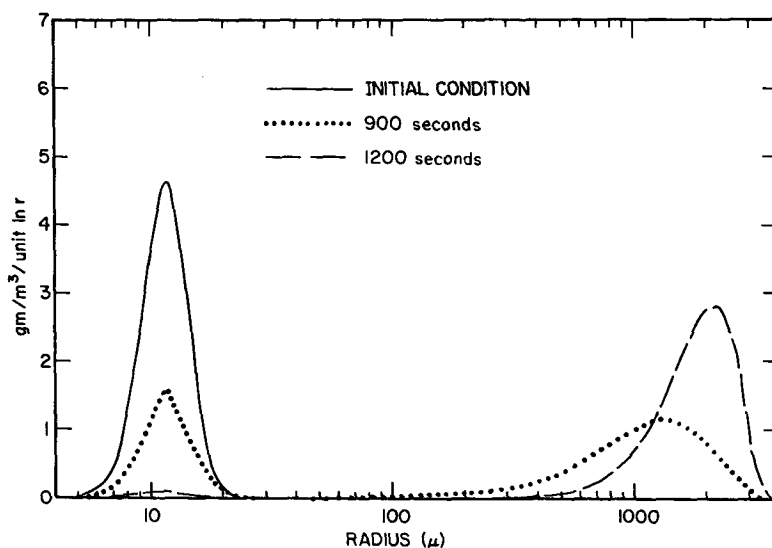


FIG. 7. As in Fig. 5 for a cloud droplet distribution having the same number concentration as that resulting from the background distribution (524.5 cm^{-3}), but the same dispersion coefficient as that resulting from the 1 km polluted distribution (0.237).

coalescence, it can be concluded that the effect of pollution on warm cloud precipitation is dependent on the pollutant particle size distribution and chemical composition. Pollutant particles which are efficient condensation nuclei can be expected to act to enhance the precipitation process, while those which are less efficient would be expected to hinder precipitation. Large effects on precipitation would not be expected from the modeled pollution. Conclusions regarding the influence of power plant effluent on precipitation development in convective clouds having an important or dominant ice phase are beyond the scope of this study and must await future research. Finally, it must be emphasized that to make realistic estimates on the effects of pollution on precipitation by the coalescence process reliable data on the size-solubility spectra of both background and pollutant particles are required.

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