A Simple Model for Droplet Size Distribution in Atmospheric Clouds

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ABSTRACT

We have used the basic probability methods of statistical mechanics to derive a droplet-size distribution function for atmospheric clouds. These methods apply to systems that have well-defined constraints, but for which microscopic processes cannot easily be followed in detail. Clouds, in their early stages of evolution, appear to be such a system. The derived expression gives the distribution as a function of the droplet volume, rather than its diameter. This agrees with a wide variety of observations on clouds for which the distribution is not bimodal, and is a convenient way to analyze data. In addition, the theory uniquely relates the width of the droplet-size distribution to the moisture content of the cloud and the concentration of condensation nuclei.

1. Introduction

Cloud formation under natural conditions involves condensation of slightly supersaturated water vapor onto condensation nuclei that are normally present in the air. (Byers, 1965; Mason, 1971; Pruppacher and Klett, 1978). The nuclei are usually smaller than 1 μm in diameter. After condensation begins, the drops grow until most of the water vapor in excess of the saturation amount has condensed. The droplets range in size from a few micrometers up to 80 μm in diameter. From this point on, the cloud evolves further, either by evaporation of droplets or by their coalescence to larger droplets that eventually fall as rain. Here we are concerned with the droplet size distribution at the end of the initial growth phase, before further evolution has begun.

2. Statistical model for droplet-size distribution

The approach used in theories of droplet growth and size distribution (Howell, 1949; Squire, 1952; Warner, 1969; Bartlett and Jonas, 1972; Fitzgerald, 1974; Lee and Pruppacher, 1977) has generally been to follow in detail the basic processes of vapor diffusion and heat conduction through the life of an individual drop. Initial conditions are assumed and then followed as a function of time. In what follows, we will describe a simple model that starts from a very different point of view. We do not follow the evolution of specific processes, but, instead, assume that conditions are complicated and cannot be followed in detail. As initial condition we assume only that there are N condensation nuclei per unit volume, and a given supersaturation, that we express as the total volume V of condensible liquid water. The average drop volume is then $\langle v \rangle = V/N$. We then seek the most probable way to distribute the volume V of liquid among the N droplets. This will lead to a distribution function of the form

$$n(v) = (N/\langle v \rangle)\exp(-v/\langle v \rangle), \quad (1)$$

where $n(v)dv$ is the number of droplets with volumes in the range v to $v + dv$. This differs in form from earlier distribution functions derived for this problem which were expressed as a function of droplet diameter rather than volume.

In a cloud, during the early stages of droplet growth, several factors may combine to produce a complex condition in which it is impossible to follow the details of droplet formation. Nuclei are randomly spaced; there is rapid air movement that may be turbulent; temperature changes arise, on a local scale from the heat of condensation of the vapor, and on a larger scale, through adiabatic cooling of uplifted air. We have used a model that neglects all these details during the process of droplet nucleation and growth. As initial conditions we take only the values of N and V. We then seek the most probable way to distribute the volume V of condensible water among the N nuclei. This is similar to the fundamental problem in statistical mechanics, where one seeks the most probable way to distribute a given total energy among a given number of molecules. One does not follow the details of the motion, but, instead, uses a probability analysis to arrive at the Boltzmann distribution. We will also use a probability analysis, as it has been used in elementary derivations of the Boltzmann distribution. However, we stress that we are making a probability calculation only, not a statistical thermodynamic calculation. The cloud droplet system is not in stable thermodynamic equilibrium and cannot be so treated. In Cohen and Turnbull (1959) a similar statistical method has been applied to a very different problem.

Our basic assumption, from which all our results derive, is that all droplet sizes physically allowed by the constraints of the system have equal *a priori* probability.

Our derivation of the most probable distribution

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closely follows the standard treatments (ter Haar, 1954) for the velocity distribution of molecules in an ideal gas. We need only those portions of the treatment, however, that deal with probability, not those that are concerned with thermodynamic equilibrium. We divide the space of possible droplet volumes into a number of cells Z_i, each cell being a volume element dv. We then distribute the actual volumes of the N droplets in such a way that the *a priori* probability for a droplet volume falling into a cell Z_i will be proportional to Z_i. The probability $W(N_i)$ that N_1 droplets will have volumes falling in Z_1, N_2 in Z_2, etc., is defined as the fraction of all possible arrangements where this particular distribution is realized, i.e.,

$$W(N_i) = CN! \prod_i \frac{Z_i^{N_i}}{N!}, \tag{2}$$

where C is a (disposable) normalization constant. We seek the desired most probable distribution by finding the maximum $W(N_i)$ subject to the constraints

$$\sum_i N_i = N, \tag{3}$$

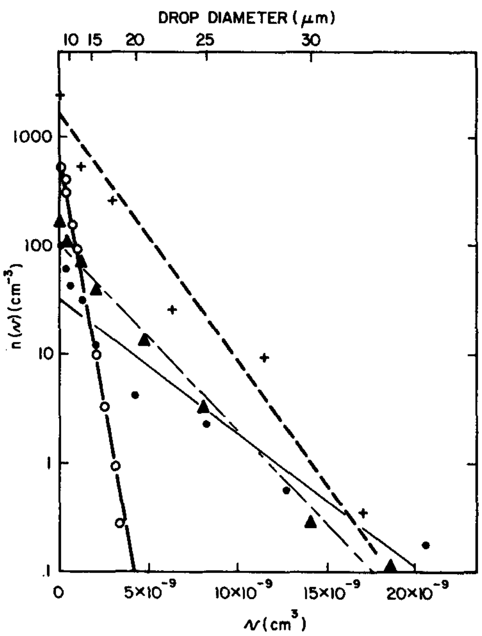

FIG. 1. Measured droplet size distributions for several clouds. Solid circles: fair weather cumulus clouds (Weickmann and aufm Kampe, 1953); crosses: cumulus clouds (Battan and Reitan, 1957); triangles: summer convective cloud over Montana (Hobbs *et al.*, 1980), UC-98; open circles: polluted stratocumulus cloud (Peuschl *et al.*, 1981). According to our model the straight line should have slope $(-1/\langle v\rangle)$. The data from (Weickmann and aufm Kampe, 1953) allow $\langle v\rangle$ to be determined experimentally. Liquid water content = 1.0 g m^{-3}; droplet concentration = 302 cm^{-3}; $\langle v\rangle = 3.3 \times 10^{-9}$ cm^3. The line connecting the solid circles has the slope $(-1/\langle v\rangle)$. For Figs. 1-3, along the vertical axis, we show $n(v)$ defined such that $n(v)$ is the number of droplets per cubic centimeter of air having droplet volumes in the range v to $v + dv$.

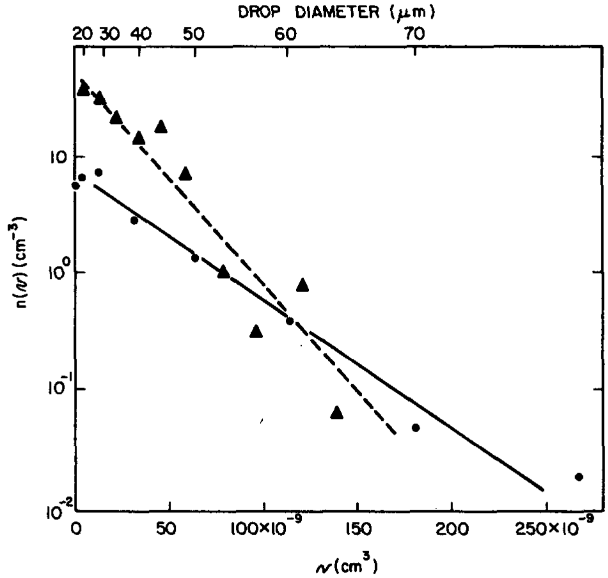

FIG. 2. Solid circles: tropical cumulus clouds over the Gulf of Mexico (Battan and Reitan, 1957); triangles: summer convective clouds over Montana (Hobbs *et al.*, 1980) UW-74. The range of droplet sizes is considerably larger than that for the data of Fig. 1. The mean droplet size is larger and the slope of the line correspondingly smaller.

$$\sum_i v_i N_i = V, \tag{4}$$

where v_i is the droplet volume corresponding to occupancy of cell Z_i. Following the usual practice we maximize the logarithm of W subject to the restrictions [Eqs. (3) and (4)] using Lagrange's method of undetermined multipliers. The resultant most probable distribution is

$$N_i = Z_i e^{-\lambda v_i} e^{-\mu}, \tag{5}$$

where $\mu - 1$ and λ are the multipliers for conditions (3) and (4), respectively. For a continuous distribution over volume we have

$$n(v)dv = e^{-\lambda v} e^{-\mu} dv. \tag{6}$$

Using Eqs. (3) and (4), the quantities λ and μ can be determined, giving the final form for the most probable distribution of droplet volumes:

$$n(v) = \frac{N}{\langle v\rangle} e^{-v/\langle v\rangle}. \tag{7}$$

We expect Eq. (7) to apply for the range of droplet sizes normally treated in cloud physics measurements, i.e., for diameters > 1 μm. We do not expect it to apply for very small values of v. For very small droplets the size of the nucleus can be a significant fraction of the droplet volume and surface tension effects become significant. Neither of these is included in our treatment.

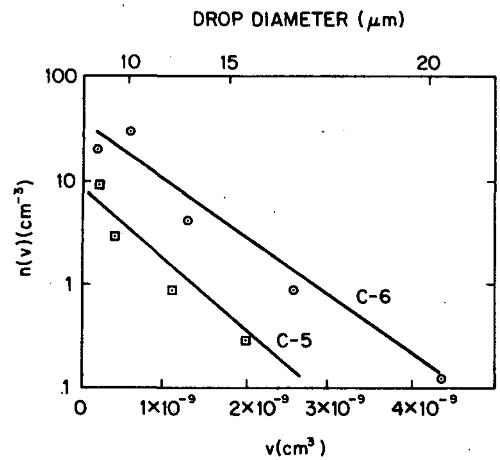

FIG. 3. Droplet size spectra for a shallow non-precipitating maritime stratus cloud over the Pacific (Ryan *et al.*, 1972). The legend c-5 and c-6 refers to a figure in reference 1 in which the data were presented. The numbers 5 and 6 indicate that the spectra were determined 5 and 6 s after cloud penetration. At later times the droplet size distribution for the cloud became bimodal.

3. Comparison with experimental data

Figs. 1–3 show comparisons of our model with published experimental data. Fig. 1 shows data from different sources for fair weather cumulus clouds. We have plotted $n(v)$ as a function of the drop volume v. We see that the data conform well to a straight line on this plot. This confirms that the model is suitable for representing data of this kind. The slope of the line is determined by the average droplet volume $\langle v \rangle$. For the data shown by the solid circles (Weickmann and aufm Kampe, 1953) $\langle v \rangle$ was determined as part of the measurement, giving us an experimental value for slope of the line according to our model. For the data represented by the solid points, the solid line has the experimental slope, derived from $\langle v \rangle$.

Fig. 2 shows data (Battan and Reitan, 1957) for tropical cumulus clouds over the Gulf of Mexico, and for summer convective clouds over Montana (Hobbs *et al.*, 1980). The range of droplet sizes and the mean size are considerably larger in this case. Fig. 3 shows data for a non-precipitating maritime stratus cloud over the Pacific (Ryan *et al.*, 1972). In this case, the average droplet sizes are smaller than for the clouds shown in Figs. 1 and 2, but, still, the size distribution has the same form.

4. Conclusion

We conclude that a simple general model, based on probability considerations alone, gives a good description of representative data for droplet size distributions on clouds. We have confined our attention to simple cases. Bimodal (Skhirtladze, 1980) and other more complicated distributions are beyond the scope of this study.

REFERENCES

Bartlett, J. T., and P. R. Jonas, 1972: On the dispersion of the droplet sizes of droplets growing by condensation in turbulent clouds. *Quart. J. Roy. Meteor. Soc.*, **98**, 150–164.

Battan, L. J., and C. H. Reitan, 1957: *Artificial Stimulation of Rain.* Pergamon Press, 184 pp.

Byers, H. R., 1965: *Elements of Cloud Physics.* University of Chicago Press, 109–118.

Cohen, M. H., and D. Turnbull, 1959: Molecular transport in liquids and glasses. *J. Chem. Phys.*, **31**, 1164–1169.

Fitzgerald, J. W., 1974: Effects of aerosol composition on cloud droplet size distribution. A numerical study. *J. Atmos. Sci.*, **31**, 1358–1367.

Hobbs, P. V., M. K. Politovich and L. F. Radke, 1980: The structures of summer convective clouds in eastern Montana. 1—Natural clouds. *J. Appl. Meteor.*, **19**, 645–663.

Howell, W. E., 1949: The growth of cloud drops in uniformly cooled air. *J. Meteor.*, **6**, 134–149.

Lee, I. Y., and H. R. Pruppacher, 1977: Comparative study on growth of cloud drops by condensation using an air parcel model with and without entrainment. *Pure Appl. Geophys.*, **115**, 523–534.

Mason, J. B., 1971: *The Physics of Clouds*, 2nd ed. Oxford University Press, 122–152.

Pruppacher, H. R., and J. D. Klett, 1978: *Microphysics of Clouds and Precipitation.* D. Reidel, 225–281.

Pueschl, R. F., E. W. Barrett, D. L. Wellman and J. McGuire, 1981: Cloud modification by man-made pollutants—effects of a coal-fired power plant on cloud drop spectra. *Geophys. Res. Lett.*, **8**, 221–224.

Ryan, R. T., H. H. Blau, P. C. Thuna and M. L. Cohen, 1972: Cloud microstructure as determined by an optical cloud particle spectrometer. *J. Appl. Meteor.*, **11**, 149–156.

Skhirtladze, G. I., 1980: Rezultaty izmereniia spektrov kapel v kuchevykh oblakakh. *Fiz. Atmos. Okeana*, **16**, 65–72.

Squires, P., 1952: The growth of cloud drops by condensation. 1—General characteristics. *Aust. J. Sci. Res.*, **5**, 59–86.

ter Haar, D., 1954: *Elements of Statistical Mechanics.* Reinhardt and Co., 22–25.

Warner, J., 1969: Micro-structure of cumulus cloud. 2—Effect on droplet size distribution of cloud nucleus spectrum and updraft velocity. *J. Atmos. Sci.*, **26**, 1272–1282.

Weickmann, H. K., and H. J. aufm Kampe, 1953: Physical properties of cumulus clouds. *J. Meteor.*, **10**, 204–211.