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Model for Time-dependent Raindrop Size Distributions; Application to the Washout of Airborne Contaminants

Abstract: We have developed a model for computing time-dependent behavior in raindrop size distributions for a variety of initial conditions. The model permits the inclusion of a spatially varying atmospheric profile and takes evaporation and coalescence of raindrops into account on a dynamic basis. We have applied this model to the washout process by rain, whereby small airborne particles are scavenged below a rain cloud. In comparing our results with those of previously published time-independent models, we found that the time-dependent effects greatly altered washout at lower elevations.

Introduction

There are two methods by which small airborne particles can be removed from the atmosphere and returned to the ground by precipitation: 1) cloud scavenging, known as rainout, in which particles mix with water droplets and ice crystals in the clouds, and 2) scavenging below the cloud level by falling raindrops, snowflakes, etc., called washout. Only washout by rain is considered here.

Previously published models of washout have assumed that the raindrop size distribution and the atmospheric conditions are the same at ground level as at the cloud base and do not vary with time. In this model we include a spatially varying atmospheric profile and take into account the evaporation and coalescence of raindrops, effects which may greatly alter the washout at lower elevations. In modeling a rainstorm, we assume that the size distribution of raindrops at any position $\mathbf{r}$ and time t can be characterized by a continuous function $N(a,\mathbf{r},t)$ such that $N da$ is the number of drops per unit volume of air in the radial interval a to $a + da$. Several authors [1] have studied the modification (due to evaporation and coalescence) of this distribution function, assuming either one-dimensional steady state conditions or spatially homogeneous conditions. In either case, the distribution histories can be computed by numerical integration of the equations that describe the coalescence and evaporation processes.

Neither steady state nor spatially homogeneous conditions are usual in nature. Strong variations in raindrop concentration often occur in both space and time. Within a sample of raindrops collected on the earth's surface, one may find drops that left the cloud base as much as ten minutes apart. Since significant changes may occur at the cloud base during such a time span, it is important to include both space and time dependence in modeling the coalescence and evaporation processes for a falling raindrop size distribution calculation.

We have chosen a model that includes radius of the raindrop, altitude, and time as independent variables in order to obtain a more realistic simulation of natural rain. The basic assumptions included are that

1. the atmosphere is constant in time and one-dimensional in that the ambient temperature, pressure, and relative humidity are functions of only the altitude, and vertical and horizontal winds are assumed zero;
2. the raindrops fall vertically at their terminal velocities, and their water vapor and heat transfer rates are governed by the differential equations given by Best [1(c)]; and
3. the transient evaporation and coalescence processes are considered simultaneously.

A numerical method was developed to determine the raindrop size distribution function $N(a,z,t)$ and the raindrop temperature $T(a,z,t)$ for radii, altitudes, and times of interest, assuming that one can specify

1. the temperature, pressure, and relative humidity profiles of the atmosphere [$\theta(z)$, $p(z)$, and $f(z)$, respectively] and

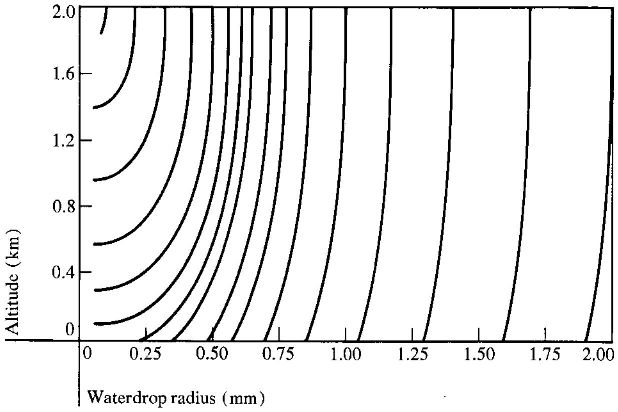


Figure 1 Evaporation histories for 16 water drops.

2. the raindrop size distribution function and raindrop temperature at the cloud base [$N(a,z_{cb},t)$ and $T(a,z_{cb},t)$, respectively].

With these assumptions we can define a scavenging rain process. When the dynamic (in the sense of coalescence and evaporation) drop falls through the atmosphere and strikes an airborne particle, we assume that the target efficiency and collision efficiency are identical. Every particle hit by a drop remains with that drop until the drop evaporates or hits the ground. Whenever a drop evaporates before reaching the ground, it releases the particle into the air at the height at which evaporation occurs. Agglomeration of particles within the raindrop is ignored. We also assume that no change in drop diameter or terminal velocity occurs at the time of capture of a particle. Any drop small enough to experience an appreciable diameter change on coalescence would have a low target efficiency and, furthermore, would undoubtedly evaporate before reaching the next lower level (within the constraints of the numerical model as described below). Thus the evaporation and coalescence equations are independent of whether the drop contains a particle.

Evaporation of a single raindrop

The evaporation of a falling waterdrop is determined by the rates of transfer of heat and water vapor between the drop surface and the surrounding air. The molecular diffusion of vapor and of heat for an evaporating waterdrop in a (stagnant) medium can be expressed by the following equations:

$$\frac{dm}{dt}=\frac{-4\pi Da}{R_w}\left[\frac{P_s(T)}{T}-\frac{fP_s(\theta)}{\theta}\right] \tag{1}$$

and

$$dH/dt = 4\pi ka(\theta - T), \tag{2}$$

where m is the mass of the drop, D is the diffusivity of water vapor in air, R_w is the gas constant for one gram of water vapor, $P_s(T)$ and $P_s(\theta)$ are the saturation vapor pressures at temperatures T and θ, respectively, f is the relative humidity, θ is the ambient air temperature, T is the surface temperature of the drop, H is the heat received by the drop through conduction, k is the thermal conductivity of air, and a is the radius of the drop.

The rate dQ/dt at which heat is received by a drop is equal to the algebraic sum of the rates at which heat is transferred to the drop by conduction, evaporation, and radiation:

$$dQ/dt = \frac{4}{3}\pi a^3 \rho C_p dT/dt$$

$$= G_1 L dm/dt + G_2 dH/dt + 4\pi a^2 \epsilon\sigma(\theta^4 - T^4). \tag{3}$$

Here L is the latent heat of vaporization, ϵ is the Stefan-Boltzmann constant, C_p is the specific heat of liquid water at constant pressure, and σ is the emissivity of the waterdrop (taken as unity). If there is significant relative motion between the drop and the surrounding medium, ventilation affects the rates of diffusion of heat and vapor. This effect has been taken into account through the empirical ventilation coefficients, G_1 for mass transfer and G_2 for heat transfer, as defined by Kinzer and Gunn[2].

Using V, the terminal velocity of the drop, which is a function of the drop size, and $\alpha = dT/dz$, the height variation of the temperature of the drop, we can replace dT/dt by αV. The heat balance equation (3) then reduces to

$$\alpha a^2 V\rho C_p = 3k(\theta - T)\ G_2 - \frac{3DL}{R_w}\ G_1\left[\frac{P_s(T)}{T}-\frac{fP_s(\theta)}{\theta}\right]$$

$$+ 3a\sigma(\theta^4 - T^4). \tag{4}$$

We can also write Eq. (1) with ventilation in the form

$$\frac{da^2}{dz}=\frac{-2DG_1}{R_w V\rho}\left[\frac{P_s(T)}{T}-\frac{fP_s(\theta)}{\theta}\right]. \tag{5}$$

Equations (4) and (5) are the two fundamental equations that we integrate numerically to determine a and T as functions of z for a specified atmosphere. [The numerical Runge-Kutta method used to solve Eqs. (4) and (5) is explained in detail in Appendix A.] Figure 1 shows sixteen individual raindrop histories computed in this manner. The atmospheric conditions for this example are $T = 15°C$, $f = 100$ percent relative humidity, and $p = 700$ mbar at the cloud base and $T = 35°C$, $f = 25$ percent relative humidity, and $p = 898$ mbar at ground level.

Evaporation and coalescence of a distribution of drops

• *Steady state evaporation*

We sample the given continuous raindrop size distribution at the cloud base (Fig. 2) at an integral number I of selected radii and assume that a set of I adjacent rectangles provides an adequate representation of the distribution. (We postpone temporarily the questions of how narrow the rectangles must be for an adequate approximation and whether unequal spacing of the sample points might be advantageous.) The sample radii and rectangles (hereafter called "bins") are numbered from largest to smallest, as shown in Fig. 2. The bin widths are defined by

$$\Delta a_1 = (a_1 - a_2),$$

$$\vdots$$

$$\Delta a_i = \tfrac{1}{2}(a_{i-1} - a_{i+1}),$$

$$\vdots$$

$$\Delta a_I = \tfrac{1}{2}(a_I + a_{I-1}); \qquad i = 2,3, \cdots, I-1. \tag{6}$$

For the special case in which the radii are sampled at uniform increments, all of the bin widths are equal. The bins change in position and size on the a axis (abscissa) as the drops evaporate.

We next compute an individual raindrop evaporation history for each of the sampled radii, using the Runge-Kutta method described by Abraham and TeSelle[3] (see Appendix A). This calculation yields the radius a, terminal velocity V, exposure time τ, and temperature T of each bin as functions of altitude.

By definition, each drop stays in its own bin and a continuity relation holds for the number of drops X_i per unit volume of air in each bin except the smallest bin, which is being depleted by evaporation. (An approximate method for treating the exceptional bin that suffers depletion through evaporation is given in Appendix B.) For other than these exceptional cases, we have from Eq. (B1) the steady state continuity relation

$$V_i dX_i/dz + X_i dV_i/dz = 0, \qquad i = 1,2, \cdots, I-1, \tag{7}$$

where $X_i = N_i\,\Delta a_i$. Separating variables and integrating from the cloud base to another altitude, we find that

$$X_i(z) = X_i(z_{\rm cb})\; V_i(z_{\rm cb})/V_i(z), \qquad i = 1,2, \cdots, I-J, \tag{8}$$

where J is the number of bins that suffer depletion of their populations due to complete evaporation. Writing this equation in terms of the distribution function, we obtain

$$N_i(z) = N_i(z_{\rm cb}) \frac{V_i(z_{\rm cb})}{V_i(z)} \frac{\Delta a_i(z_{\rm cb})}{\Delta a_i(z)}, \qquad i = 1,2, \cdots, I-J. \tag{9}$$

Equation (9) describes the modification of a raindrop size distribution due to evaporation under steady state conditions.

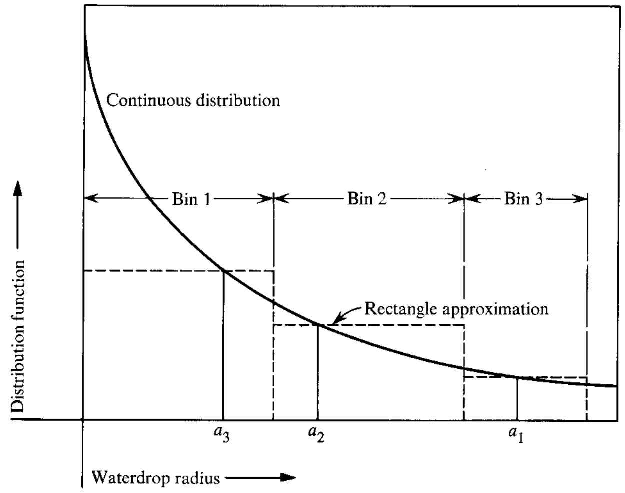


Figure 2 Continuous size distribution sampled at selected radii.

• *Transient evaporation*

The extension of the steady state solution to the time-dependent case consists merely of introducing an appropriate time delay in the process. For this purpose it is convenient to label altitudes from the cloud base downward, since that is the direction of numerical integration; thus altitude z_n is greater than z_{n+1}. If we assume that it requires τ_i seconds for a drop in bin i to fall from altitude z_n to altitude z_{n+1}, the fall time is defined by

$$\tau_i = \int_{z_n}^{z_{n+1}} d\xi/V_i(\xi), \qquad i = 1,2, \cdots, I. \tag{10}$$

The distribution function at altitude z_{n+1} at time t depends on the distribution function at altitude z_n at time $t - \tau_i$. This time delay is the only feature of the transient problem that is not accounted for in the steady state case.

The transient solution for the number of drops in each bin is

$$X_i(z_{n+1},t) = X_i(z_n, t-\tau_i)\frac{V_i(z_n)}{V_i(z_{n+1})},$$

or

$$N_i(z_{n+1},t) = N_i(z_n, t-\tau_i)\,\frac{V_i(z_n)}{V_i(z_{n+1})}\,\frac{\Delta a_i(z_n)}{\Delta a_i(z_{n+1})},$$
$$i = 1,2, \cdots, I-J. \tag{11}$$

The solution (11) must satisfy the time-dependent continuity relation (except for the exceptional cases already mentioned)

$$\partial X_i/\partial t + V_i \partial X_i/\partial z + X_i dV_i/dz = 0, \quad i = 1,2, \cdots, I-J.$$

This is shown to be the case in Appendix C.

• *Steady state coalescence*

Coalescence takes place when two falling drops collide and unite, giving rise to a larger drop. Under steady state

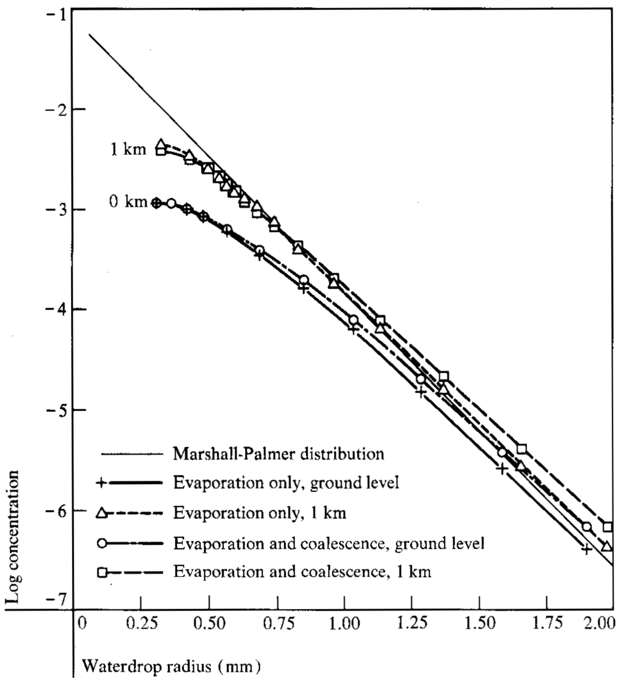


Figure 3 Transient modification of the raindrop distribution function due to time-varying concentration at the cloud base (2 km), evaporation, and coalescence. The distribution function was initialized at the cloud base using the Marshall-Palmer distribution law with a precipitation rate of 3.5 mm/h and is shown at two altitudes after eight minutes of rain.

conditions, the coalescence process may be described by either a single nonlinear partial differential equation or a system of nonlinear ordinary differential equations, depending on whether the size distribution is continuous or discrete[Hidy and Lilly, 1(b)]. If we continue to use drops at sampled radii to represent the distribution, the system of ordinary differential equations is appropriate.

The number C_{ij} of collisions per unit volume of air and unit time between drops with radii a_i and a_j is

$$C_{ij} = \pi(a_i + a_j)^2 \mid V_i - V_j \mid X_i X_j, \tag{12}$$

if the collection efficiency is taken as unity. The rate of depletion of drops with radius a_j is

$$dX_j/dz = -\sum_{i=1}^{I} C_{ij}/V_j. \tag{13}$$

The combined drop will have radius $a_c = (a_i^3 + a_j^3)^{1/3}$ and terminal velocity V_c after accumulation. The accumulation of drops with this radius as a function of altitude is determined by the conservation of mass equation

$$dX_c/dz = (a_i^3 C_{ji} + a_j^3 C_{ij})/a_c^3 V_c. \tag{14}$$

This rate of accumulation may be divided between the nearest neighbor bins (radii a_{k+1} to a_c and a_c to a_k) by the equations

$$\frac{dX_{k+1}}{dz} = \left(\frac{a_k^3 - a_c^3}{a_k^3 - a_{k+1}^3}\right)\frac{dX_c}{dz}. \tag{15}$$

and

$$\frac{dX_k}{dz} = \left(\frac{a_c^3 - a_{k+1}^3}{a_k^3 - a_{k+1}^3}\right)\frac{dX_c}{dz}. \tag{16}$$

The sum of the rates of depletion and accumulation for each bin yields the net rate of change of concentration for that bin.

- *Transient coalescence*

The coalescence rate at any radius depends only on the distribution function and the drop velocities at the time in question. Under these circumstances, we can treat time as a parameter in the rate equations and replace the ordinary derivatives in Eqs. (13) through (16) with partial derivatives (see Appendix D).

- *Comparison of transient and steady state equations*

We have found that the fundamental equation describing evaporation in a raindrop size distribution differs from the steady state equation only by a simple time delay. Since this time delay is easily incorporated into a computer solution, it is only slightly more difficult to calculate distribution histories with three independent variables than with two such variables. Implementation of the three-variable program results in Fig. 3, in which the logarithm of the concentration of drops is plotted as a function of drop radius. The atmospheric conditions assumed are 15°C, 100 percent relative humidity, 700 mbar at the cloud base and 35°C, 25 percent relative humidity, 898 mbar at ground level.

The distribution function at the cloud base at time zero was initialized by using a Marshall-Palmer distribution law[4] with a precipitation rate of 3.5 mm/h [Fig. 3(a)]. After a time of 8 min, the calculated modification to the distribution function is shown for cases considering (b) only evaporation effects and (c) evaporation and coalescence effects. The curves for altitudes of 1 and 2 km intersect at a drop radius of 1.25 mm. This is due to the coalescence of drops as they fall to the 1-km level.

Washout

As an illustration of the raindrop spectrum model, we considered the washout of large aerosol particles from the atmosphere by rain. For convenience, we limited the computation to particles for which the Langmuir efficiencies[9] may be used.

Our model requires that all of the airborne particles be of uniform size and density. The particles should be small enough (20 μm or less in diameter) that their terminal velocity is negligible, and large enough (at least 2μm) that the possible electric charges on drops and particles may be ignored. We have assumed that the particles have the same density as water. If they do not, a good approximation can be made by replacing the radius a by $a(\rho_1/\rho_2)^{1/2}$, where ρ_1 is the density of the particles and ρ_2 is the density of water. The value of $a(\rho_1/\rho_2)^{1/2}$ should be between 2 and 20 μm.

If no falling drop that originated at the cloud base disappears (evaporates completely) before reaching the ground, every airborne particle that is captured is actually removed from the atmosphere. However, if drops do disappear, any particles that they have picked up are released back into the atmosphere at the height at which these drops evaporate. First consider the case in which none of the drops disappears. Then at each height interval the contamination χ_i left at time t_i is[5]

$$\chi_i = \chi_{i-1} \exp[-(t_i - t_{i-1})\,\Lambda]. \tag{17}$$

The washout coefficient Λ is computed as

$$\Lambda = \int_0^\infty NVEA\,da, \tag{18}$$

where for radius a, N represents the concentration (drops/cm^4) of the raindrops in the air at that height level; V is the terminal velocity (cm/min) of the drops; E is the collision (target) efficiency of drops against particles of prescribed diameter (the program uses a linear interpolation of the Langmuir efficiencies); and A is the cross-sectional area of the drops.

Now consider the case in which some of the drops whose cloud-base radius is a disappear when they reach the height of a given bin. The contamination at this level at time t_i is increased by $\bar{\chi}_i$, where $\bar{\chi}_i$ is the contamination carried by these evaporating drops. For the moment, we ignore the effect of drop coalescence. The increment $\bar{\chi}_i$ is the sum over all heights z from the cloud base to the height z_h of the concentrations of contaminants that are picked up by these drops between times t_{i-1} and t_i. The time t_i is a function of z and is the time at which a drop with initial radius a was at height z if it reached height z_h between times t_{i-1} and t_i. Thus we have the average values

$$\bar{\chi}_i = \bar{\chi}(t_i,z_h) = \sum_{z<z_h} \frac{NVEA(\bar{t}_i,z)}{\Lambda(\bar{t}_i,z)} [\chi(\bar{t}_i,z) - \chi(\bar{t}_{i-1},z)]. \tag{19}$$

We next investigate the effect of coalescence on $\bar{\chi}_i$. Consider that a drop which initially had radius a coalesces with another drop. Since coalescence increases the final radius, the raindrop will not evaporate at height z_h and hence will not release any contaminant into the air at that height. To take coalescence into account, we simply revise the above calculation of $\bar{\chi}_i$ to

$$\bar{\chi}_i = \sum_{z<z_h} \frac{NVEA(\bar{t}_i,z)}{\Lambda(\bar{t}_i,z)} [\chi(\bar{t}_i,z) - \chi(\bar{t}_{i-1},z)]\;\Phi(\bar{t}_i,z), \tag{20}$$

where $\Phi(t,z)$ is a correction factor due to coalescence, namely, the fraction of drops that coalesce.

Once the release of contaminant begins to take effect, the total concentration of contaminant at a given height at time t_i is

$$\chi_i = \bar{\chi}_i + \chi_{i-1} \exp[-(t_i - t_{i-1})\,\Lambda], \tag{21}$$

instead of Eq. (17).

As has been mentioned, our model uses a discrete set of raindrop sizes (bins) rather than a continuous distribution. Now consider the computational formulation of the washout coefficient Λ. At any particular elevation, the parameters V, E, and A are functions of only the number of bins, NB. The drop concentration N depends on the time as well as on the particular bin. The concentration of contaminant left at time t_i depends on the distribution of raindrops throughout the time interval t_{i-1} to t_i. This distribution is computed only at specified times—the same times for which the washout results are to be computed. An average of N at times t_i and t_{i-1} is used as an estimate of N over that time interval. Once this estimate is made, we can compute Λ for the time interval as

$$\begin{aligned}\Lambda &= \int_0^\infty NVEA\,da \\ &= \sum_{NB=1}^{N} \tfrac{1}{2}[N(t_i) + N(t_{i-1})]\; VEA\Delta(NB),\end{aligned} \tag{22}$$

where $\Delta(NB)$ represents the width of the bin at the height of interest.

To reformulate Eq. (20) for the computer program, we took the height z in 50-m intervals. Since N, Λ, χ, and Φ are calculated only at specified times, we do not necessarily know their values at t_i and must use whichever of the specified times is closest to, but not greater than, t_i. Furthermore, in the model the drops may evaporate only at discrete heights whereas, in reality, the drop evaporation heights should be a continuum. We approximate the continuum by distributing the contaminant throughout the entire bin if the drop evaporates in that bin.

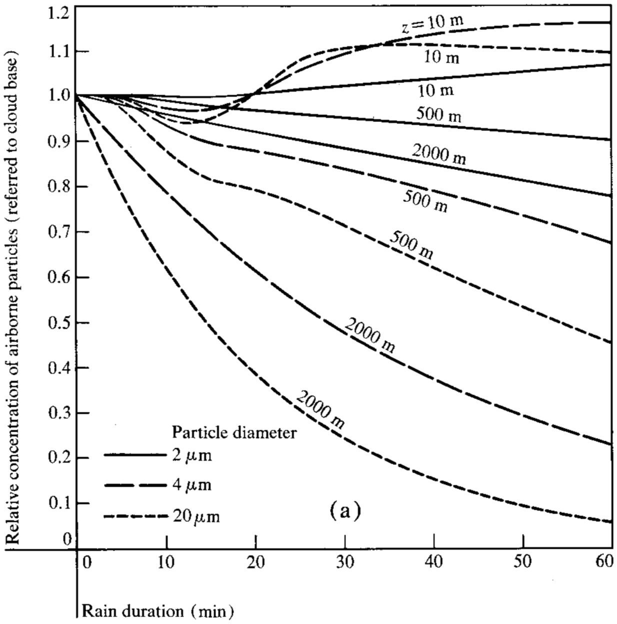
Results

Our calculations were tested using Best's rain spectra data[1(c)]. We compared the washout at the cloud base (taken as 600 m, which is the height at which the spectra were measured) for two rainfall intensities and three particle sizes with the washout predicted by Chamberlain[6], who also based his calculations on Langmuir's collision efficiencies and Best's spectra. Using Table 1, one sees that the agreement between our computations and Chamberlain's predictions is excellent. Since Chamberlain did not take the effect of evaporation into account, comparisons at lower elevations were not made.

Several computations were made to determine how the concentration of contaminant depends on factors such

Table 1 Comparison of washout calculations at the cloud base (600 m).

Rainfall intensity (mm/h)	Particle diameter (μm)	Time (min)	Relative concentration: Present work	Relative concentration: Chamberlain (Ref. 6)
4	4	0	1.00	1.00
		10	0.73	0.72
		30	0.38	0.37
		60	0.15	0.14
0	20	0	1.00	1.00
		10	0.61	0.60
		30	0.22	0.22
		60	0.050	0.047



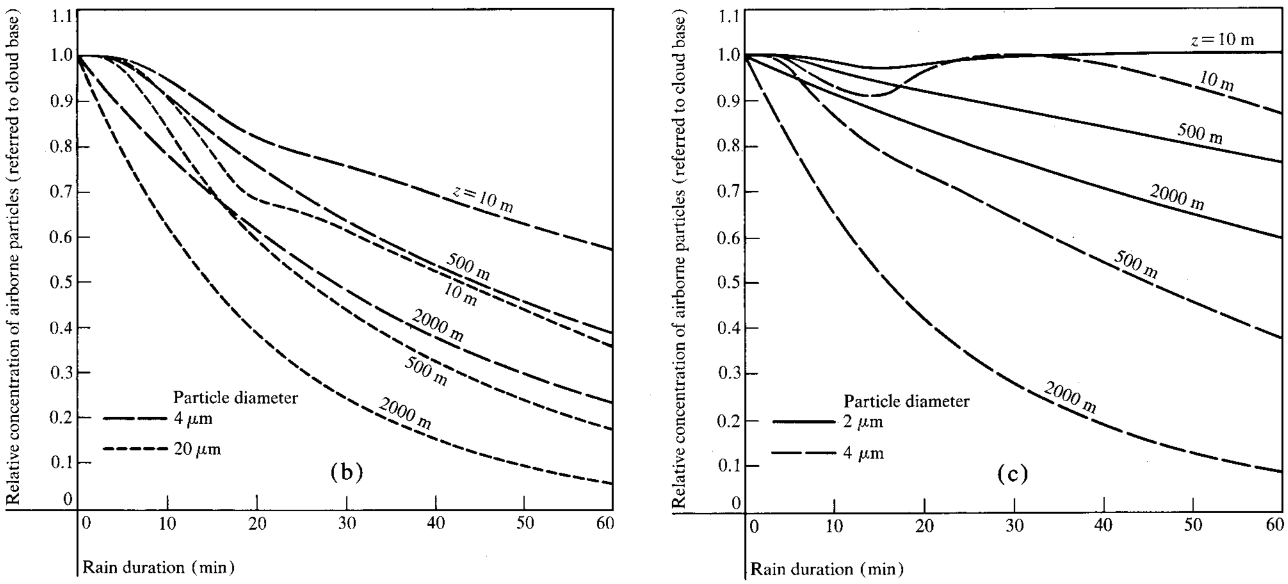


Figure 4 (a) Concentration of contaminant particles in air as a function of rain duration. Rainfall intensity at the cloud base (2 km) is 3.5 mm/h, ambient temperature 15°C, relative humidity 100 percent, pressure 700 mbar; at ground level, ambient temperature 35°C, relative humidity 25 percent, pressure 898 mbar. Note that few small particles get washed out, only 17 percent after 60 min, and that the concentration is still increasing at ground level after one hour. (b) Humidity at ground level increased to 75 percent, other conditions the same as in (a); washout is increased. (c) Rainfall intensity at the cloud base doubled, other conditions the same as in (a); washout is increased, but even after one hour the concentration at ground level is still about the same as it was originally for the 2-μm particles.

as elevation, humidity profile, particle size, and rainfall intensity. Figure 4 shows the concentration of contaminants in the air at different altitudes over a period of time. The relation of particle size to washout can be inferred from Fig. 4(a). The comparison of 4(a) with 4(b) suggests the effect of humidity, while the comparison of 4(a) with 4(c) illustrates the expected relation to rainfall intensity.

Even though the constant ambient conditions represent an unrealistic model for a rainstorm (especially the 25 percent relative humidity case), the ambient extremes clearly demonstrate the importance of these conditions on the washout phenomenon. A logical extension of this model would be the updating of the ambient conditions during the rain.

Initially, the concentration at all altitudes was set at 1.00 (arbitrary units). At the cloud base, washout begins immediately, but at lower altitudes there is a delay until the first large drops arrive and begin to collect the airborne particles. There is another delay before the first small drops (whose terminal velocity is less) reach the height at which they evaporate and release the particles they carry. Inspection of Fig. 4 shows that less contamination is washed out at low levels than at high levels.

There are two reasons for this. First, all drops decrease in size as they fall, and hence their collision efficiencies are decreasing; that is, they pick up fewer and fewer particles as they descend. Second, those particles that evaporate completely only transport particles from higher elevations to lower ones. In some cases this second effect completely overshadows the scavenging effect of larger drops, and the concentration of particles in the air may actually be increasing with time at the lower levels. However, given enough time, or great enough rainfall intensity, the contaminant concentration eventually decreases.

As an example, consider the concentration of 20-μm airborne particles at a height of 10 m in Fig. 4(a). The effect of washout is observable only after a time of approximately 5 min because of the delay of the onset of precipitation. Washout proceeds for about 5 min, after which the rate of change of concentration begins to decrease. The concentration of particles starts to increase at 12 min. This is due to particles from higher levels being carried to lower levels by evaporating raindrops. Particulate concentration reaches a maximum at 40 min and decreases thereafter.

The humidity profile from ground level to the cloud base was assumed to be linear. The calculations for Fig. 4(b) were based on higher humidity at all elevations than the case in Fig. 4(a). Thus 4(b) indicates that the drops evaporate relatively slowly and fewer drops disappear completely. Since large drops have greater collision efficiencies than small ones, more particles are picked up. Also, since fewer drops disappear, fewer drops release particles at lower elevations. These effects contribute to the removal of a greater percentage of contamination from the air for the 75 percent relative humidity case.

Because the collection efficiency for all raindrop sizes is greater for larger contaminant particles, a greater percentage of large particles is removed during a rainstorm. Also, the greater the rainfall intensity, the faster the particles are washed out. At the cloud base, the washout coefficient is roughly proportional to the rainfall intensity, although this relation does not hold at lower levels where contamination is being released by evaporating drops.

Summary

This model has successfully simulated the rain-washout process whereby small airborne particles are removed from the atmosphere and returned to the ground by precipitation. The model includes a spatially varying atmospheric profile and takes evaporation and coalescence of raindrops into account explicitly. We have assumed that the atmosphere is constant in time and one-dimensional in that the ambient temperature, pressure, and relative humidity are functions of altitude only, while vertical and horizontal winds are assumed zero. We have further assumed that the raindrops fall vertically at their terminal velocities. The model treats transient evaporation and coalescence simultaneously. It tracks a distribution of sizes of drops as a function of time. The scavenging of airborne particles by the drops is inserted as the last step in the calculation.

The results clearly show the value of computer simulation in studying the scavenging process of airborne particles. An excellent survey of this developing field can be found in the proceedings of a symposium on precipitation scavenging held at Richland, Washington, in June 1970[10].

Appendix A—Evaporation of a single raindrop

In this appendix we present the details of a computer-oriented method for finding the evaporation histories of single raindrops falling through atmospheres of nonuniform temperature and humidity.

The formulation of the appropriate equations for the evaporation of falling waterdrops presents little difficulty. However, an exact analytical solution of the equations is not possible. Best[1(c)] replaced awkward expressions in the equations by expressions having simpler forms in order to permit integration of the equations. The simpler forms were derived by evaluating the awkward expressions and fitting simpler formulas to them by empirical methods.

We use a numerical Runge-Kutta method[7] with variable step size[8]. The basic Runge-Kutta scheme for the solution of two simultaneous equations is shown below. This method is of fourth-order accuracy, since it can be shown to agree with the Taylor series expansion through the $(\Delta z_n)^4$ term.

The two differential Eqs. (5) and (4) can be rewritten as

$$\frac{db}{dz} = \frac{-2DG_1}{R_w V\rho}\left[\frac{P_s(T)}{T} - \frac{fP_s(\theta)}{\theta}\right] \tag{A1}$$

and

$$\frac{dT}{dz} = \frac{3}{bVC_p\rho}\left[kG_2(\theta - T) + \tfrac{1}{2}L\rho V\frac{db}{dz} + b^{\frac{1}{2}}\sigma(\theta^4 - T^4)\right]. \tag{A2}$$

where $b = a^2$. We make the following definitions relating the equations to the increment Δz:

$$b(z_0) = b_0 = a_0^{\ 2};$$

$$db/dz = F(z,b,T);$$

$$T(z_0) = T_0;$$

$$dT/dz = G(z,b,T);$$

$$k_0 = \Delta z F(z_i,b_i,T_i);$$

$$k_1 = \Delta z F(z_i + \tfrac{1}{2}\Delta z,\ b_i + \tfrac{1}{2}k_0,\ T_i + \tfrac{1}{2}I_0);$$

$$k_2 = \Delta z F(z_i + \tfrac{1}{2}\Delta z,\ b_i + \tfrac{1}{2}k_1,\ T_i + \tfrac{1}{2}I_1);$$

$$k_3 = \Delta z F(z_i + \Delta z,\ b_i + k_2,\ T_i + I_2);$$

$$I_0 = \Delta z G(z_i,b_i,T_i);$$

$$I_1 = \Delta z G(z_i + \tfrac{1}{2}\Delta a,\ b_i + \tfrac{1}{2}k_0,\ T_i + \tfrac{1}{2}I_0);$$

$$I_2 = \Delta z G(z_i + \tfrac{1}{2}\Delta z,\ b_i + \tfrac{1}{2}k_1,\ T_i + \tfrac{1}{2}I_1); \qquad \text{and}$$

$$I_3 = \Delta z G(z_i + \Delta z,\ b_i + k_2,\ T_i + I_2). \tag{A3}$$

The Runge-Kutta solutions[7] can then be written as

$$b_{i+1} = b_i + \frac{1}{6}\,(k_0 + 2k_1 + 2k_2 + k_3) \tag{A4}$$

and

$$T_{i+1} = T_i + \frac{1}{6}\,(I_0 + 2I_1 + 2I_2 + I_3), \tag{A5}$$

where $a_{i+1} = (b_{i+1})^{1/2}$.

An error function is used to determine whether the accuracy of the solution is within our (arbitrary) limits of allowable error and, if not, to adjust the increment size Δz for use in the next iteration. We make the additional definitions

$$k_4 = \Delta z F(z_{i+1},\ b_{i+1},\ T_{i+1})$$

and

$$I_4 = \Delta z G(z_{i+1},\ b_{i+1},\ T_{i+1}). \tag{A6}$$

The error functions are defined by[8]

$$E_1 = k_0 - 2(k_2 + k_3) + 3k_4 \tag{A7}$$

and

$$E_2 = I_0 - 2(I_2 + I_3) + 3I_4. \tag{A8}$$

To use the error functions we define an upper and a lower limit, U and L, respectively, against which the error functions are checked. If the solution is not accurate enough, the increment Δz is halved for the next iteration. If the solution is "too accurate," i.e., if the increment is smaller than necessary, then Δz is doubled for the next iteration. Each of the limits consists of an absolute term and a relative term. Thus, for a generalized solution y_i of (A1) and (A2), the logic of the error routine is

$$E < L_{\mathrm{abs}} + L_{\mathrm{rel}}\,|\,y_i\,| \;\rightarrow \Delta z = 2\Delta z,$$

$$E > U_{\mathrm{abs}} + U_{\mathrm{rel}}\,|\,y_i\,| \;\rightarrow \Delta z = \tfrac{1}{2}\Delta z, \qquad \text{and}$$

$$L_{\mathrm{abs}} + L_{\mathrm{rel}}\,|\,y_i\,| \;\leq E \leq U_{\mathrm{abs}} + U_{\mathrm{rel}}\,|\,y_i\,|$$

$$\rightarrow \Delta z \text{ unchanged.} \tag{A9}$$

Appendix B—Total depletion of bins by evaporation

At any altitude and time during the distribution history calculations, there is only one bin (the smallest one) for which the concentration does not obey the continuity relation. A typical bin structure is shown in Fig. 2. The smallest drops in this bin are "disappearing." Since there is no exact solution for the rate at which these drops disappear, we have developed an approximate treatment.

Given the concentrations in all the bins at an upper altitude, we want to determine the concentration of drops in bin i at the next lower altitude. If we ignore the depletion of the population of the smallest bin due to evaporation, i.e., if we assume that the continuity relation holds, we have

$$X_i(z-h,t) = X_i(z,t-\tau_i)\ V_i(z)/V_i(z-h). \tag{B1}$$

The actual concentration in bin i will be somewhat smaller than this value because drops are evaporating. Therefore we modify Eq. (B1) to

$$X_i(z-h,t) = X_i(z,t-\tau_i)\ \frac{V_i(z)}{V_i(z-h)}\ \frac{\Delta a_i(z-h)}{\Delta a_i(z)}. \tag{B2}$$

In terms of the distribution function, Eq. (B2) becomes

$$N_i(z-h,t) = N_i(z,t-\tau_i)\ V_i(z)/V_i(z-h). \tag{B3}$$

Thus our approximation consists of assuming that for small drops the distribution function is modified primarily by variations in the *terminal velocity*, rather than by variations in the bin width.

Appendix C—Nonequilibrium continuity equation

The nonequilibrium continuity equation, which relates the variation of raindrop concentration to terminal velocity of the drops, is

$$\left(\frac{\partial X_i}{\partial t}\right)_z + V_i\left(\frac{\partial X_i}{\partial z}\right)_t + X_i\,\frac{dV_i}{dz} = 0, \qquad i = 1,2,\cdots,I-1, \tag{C1}$$

where $X_i = X_i(z,t)$ and $V_i = V_i(z)$. A separate homogeneous equation is written for each bin since, in the absence of coalescence, there is no mass transfer between the bins.

For a drop falling from altitude z_0 to altitude z, the proposed solution of the continuity equation is

$$X_i(z,t) = X_i(z_0,t-\tau_i)\ V_i(z_0)/V_i(z), \tag{C2}$$

where

$$\tau_i(z) = \int_{z_0}^{z} \frac{d\xi}{V_i(\xi)}. \tag{C3}$$

Evaluating the partial derivatives and using $Q_i(z,t) = t - \tau_i(z)$ for convenience, we have

$$\left(\frac{\partial X_i}{\partial t}\right)_z = \left(\frac{\partial X_i}{\partial Q_i}\right)_z \left(\frac{\partial Q_i}{\partial t}\right)_z = \left(\frac{\partial X_i}{\partial Q_i}\right)_z \tag{C4}$$

and

$$\begin{aligned}\left(\frac{\partial X_i}{\partial z}\right)_t &= \left(\frac{\partial X_i}{\partial z}\right)_{Q_i} + \left(\frac{\partial X_i}{\partial Q_i}\right)_z \left(\frac{\partial Q_i}{\partial z}\right)_t \\ &= -X_i(z_0,Q_i)\,\frac{V_i(z_0)}{V_i^{\,2}(z)}\,\frac{dV_i}{dz} - \left(\frac{\partial X_i}{\partial Q_i}\right)_z \frac{d\tau_i}{dz},\end{aligned} \tag{C5}$$

and from Eq. (C3)

$$d\tau_i/dz = 1/V_i(z). \tag{C6}$$

Substituting these latter three equations into the continuity equation (C1), we have

$$\begin{aligned}\left(\frac{\partial X_i}{\partial Q_i}\right)_z - \left(\frac{\partial X_i}{\partial Q_i}\right)_z - X_i(z_0,Q_i)\,\frac{V_i(z_0)}{V_i(z)}\,\frac{dV_i}{dz} \\ + X_i(z_0,Q_i)\,\frac{V_i(z_0)}{V_i(z)}\,\frac{dV_i}{dz} = 0,\end{aligned} \tag{C7}$$

which verifies that the proposed solution (C2) conserves the total population.

Appendix D – Derivation of numerical difference equations

When transient evaporation and coalescençe effects are considered simultaneously, the equation of continuity can be written as

$$\begin{aligned}&\partial X_i/\partial t + V_i\,\partial X_i/\partial z + X_i\,dV_i/dz \\ &\quad = V_i f_i[\bar{a}(z),\overline{V}(z),\overline{X}(z,t)],\end{aligned} \tag{D1}$$

where the rate f_i of change of concentration due to coalescence in the ith bin is obtained from Eqs. (12) through (16). Bars are used on the right-hand side of Eq. (D1) to emphasize that the coalescence rate of bin i depends on the radii, velocities, and concentrations of all the bins.

Considering $\overline{V}(z)$ and $\bar{a}(z)$ as known functions, which are obtained by numerical integration of the water vapor and heat transfer differential equations for a single raindrop, we seek a solution of Eq. (D1) with the initial condition

$$\overline{X}(z_{\rm cb},t) = \overline{X}^0(t), \tag{D2}$$

where $\overline{X}^0(t)$ is an assigned value. Equation (D1) represents a first-order, nonlinear, partial differential equation which can be reduced to a first-order ordinary differential equation by a change of coordinates.

We define a new variable Γ such that the new independent variables are

$$\Gamma \equiv u(z,t) = t - \int_{z_{\rm cb}}^{z} d\xi/V_i(\xi) \quad \text{and} \quad s \equiv z. \tag{D3}$$

Using the chain rule for differentiation, we have

$$(\partial/\partial t)_z = (\partial u/\partial t)_z\,(\partial/\partial\Gamma)_s \tag{D4}$$

and

$$(\partial/\partial z)_t = (\partial/\partial s)_\Gamma + (\partial u/\partial z)_t\,(\partial/\partial\Gamma)_s, \tag{D5}$$

and we find that Eq. (D1) transforms into

$$[\partial(X_iV_i)/\partial s]_\Gamma = V_iF_i[\bar{a}(s),\overline{V}(s),\overline{X}(s,\Gamma)]. \tag{D6}$$

Since (D6) is an ordinary differential equation, it can be solved by standard techniques.

We solved Eq. (D6) numerically using difference equations much like the Runge-Kutta difference equations. The only distinction between the usual Runge-Kutta algorithm and the one used here is due to the time delays. In the difference equations, altitudes are indexed from the cloud base downward and the altitude step size is h; τ_i' is the time for a drop in bin i to fall from altitude z_n to altitude $z_n - \frac{1}{2}h$; τ_i'', from $z_n - \frac{1}{2}h$ to $z_n - h$; and τ_i''', from z_n to z_{n+1}. (Note that $\tau_i''' = \tau_i' + \tau_i''$.)

The numerical solution of Eq. (D6) from altitude z_n to altitude z_{n+1} proceeds as follows:

1. Compute the coalescence rates at altitude z_n:

$$\begin{aligned}&f_i(z_n,t) = f_i[\bar{a}(z_n),\overline{V}(z_n),\overline{X}(z_n,t)], \\ &\qquad i = 1,2,\cdots,I;\; t = t_1,t_2,\cdots,t_m,\cdots,t_M\end{aligned} \tag{D7}$$

(These same iterations on radius and time are implicit in the next four steps.)

2. Compute the concentrations and coalescence rates at altitude $z_n - \frac{1}{2}h$:

$$\begin{aligned}X_i(z_n - \tfrac{1}{2}h,t) &= X_i(z_n,t-\tau_i')\,\frac{V_i(z_n)}{V_i(z_n - \frac{1}{2}h)} \\ &\quad + \tfrac{1}{2}h\,f_i(z_n,t-\tau_i');\end{aligned} \tag{D8}$$

$$f_i(z_n = \tfrac{1}{2}h,t) = f_i[\bar{a}(z_n - \tfrac{1}{2}h),\overline{V}(z_n - \tfrac{1}{4}h),\overline{X}(z_n - \tfrac{1}{2}h,t)]. \tag{D9}$$

3. Compute the corrected concentrations and coalescence rates at altitude $z_n - \frac{1}{2}h$:

$$\begin{aligned}X_i^{\rm c}(z_n - \tfrac{1}{2}h,t) &= X_i(z_n,t-\tau_i')\,\frac{V_i(z_n)}{V_i(z_n - \frac{1}{2}h)} \\ &\quad + \tfrac{1}{2}h\,f_i(z_n - \tfrac{1}{2}h,t);\end{aligned} \tag{D10}$$

$$\begin{aligned}&f_i^{\rm c}(z_n - \tfrac{1}{2}h,t) \\ &\quad = f_i[\bar{a}(z_n - \tfrac{1}{2}h),\overline{V}(z_n - \tfrac{1}{2}h),\overline{X}^{\rm c}(z_n - \tfrac{1}{2}h,t)].\end{aligned} \tag{D11}$$

4. Compute the concentrations and coalescence rates at altitude $z_n - h$:

$$X_i(z_n - h,t) = X_i(z_n,t - \tau_i''')\,\frac{V_i(z_n)}{V_i(z_n - h)} + h\,f_i^{c}(z_n - \tfrac{1}{2}h,t - \tau_i''); \tag{D12}$$

$$f_i(z_n - h,t) = f_i[\bar{a}(z_n - h),\overline{V}(z_n - h),\overline{X}(z_n - h,t)]. \tag{D13}$$

5. Compute the corrected concentrations at altitude $z_n - h$:

$$X_i^{c}(z_n - h,t) = X_i(z_n,t - \tau_i''')\,\frac{V_i(z_n)}{V_i(z_n - h)} + \tfrac{1}{6}h[f_i(z_n,t - \tau_i''') + 2f_i(z_n - \tfrac{1}{2}h,t - \tau_i'') + 2f_i^{c}(z_n - \tfrac{1}{2}h,t - \tau_i'') + f_i(z_n - h,t)]. \tag{D14}$$

The cycle 1 through 5 is repeated after increasing the index n on altitude by one, and continues until altitude zero is reached. To begin the next cycle, set

$$X_i(z_{n+1},t) = X_i^{c}(z_n - h,t). \tag{D15}$$

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Received February 18, 1971; revised August 23, 1971

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