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NUMERICAL MODELS FOR PRECIPITATION SCAVENGING

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ABSTRACT

NUMERICAL MODELS FOR PRECIPITATION SCAVENGING

by

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Two models are developed. The first is designed for precipitation scavenging in stratiform clouds. This model incorporates heuristic approximations of diffusive attachment, impact collection and accretion, and includes consideration of particle and cloud droplet size spectra. An overriding, independently generated steady rain removes contaminated cloud droplets and contaminant particles. The conservation of contaminant mass is expressed by a set of three ordinary differential equations for which analytic solutions are derived. Rainout ratios and efficiencies are computed, and the results compare favorably with available field measurements. It is shown 1) that no single attachment rate constant or removal rate constant can adequately express the removal by rain of airborne contaminant, 2) that the contaminant concentration in the cloud air decreases exponentially with time when the processes of diffusive attachment and rainfall removal are considered, and 3) that the fraction of the air concentration of contaminant that is attached to the cloud droplets is affected by processes of particle attachment and rainfall removal, hence its temporal change shows an initial rapid rise to a maximum which is followed by a variable decrease

that depends upon the rainfall rate and the cloud water content.

The second model addresses the problem of air cleansing by a convective rain-generating system. Microphysical processes such as Brownian motion and impact collection can be neglected because the residence time in the cloud of the particles is too short for Brownian attachment, and the particles are too small for efficient impact collection by raindrops. Therefore, in this model, raindrop growth and motion in spatial contaminant concentration gradients are of major concern. Numerical solutions for raindrops growing by coalescence with cloud droplets and moving in a cylindrically symmetric motion field within a steady-state cloud are provided. The drop breakup process is formally accounted for, and, in the sub-cloud region, evaporation is considered. The cloud water and contaminant concentration are assumed to vary both with height and horizontal distance from the updraft core. A bell-shaped updraft profile is assumed. The contaminant concentrations of individual raindrops are computed. Because the updraft core is identified with the maxima of liquid water content and contaminant concentrations, (a) the coalescence growth process is most rapid in the upward passage of droplets through the cloud rather than the downward passage; (b) high concentration of contaminant tends to be associated with the larger drops formed near the core and lower concentration tends to be found in the smaller drops formed farther away from the core, and these associations produce a positive correlation between rainfall rate and contaminant concentration. Sub-cloud evaporation, on the other hand, tends to produce a negative correlation between these two parameters because it affects the small drops much more strongly than the large ones. Drops of different sizes can therefore have the same concentration, and under the assumed conditions a particular drop-size is identified with the minimum concentration.

Inasmuch as these several relationships are observed in field experiments, the present model fulfills the special constraints imposed by the field data at the same time that it represents a plausible combination of the characteristics of previous convective storm models.

CHAPTER 1

INTRODUCTION

The atmosphere is polluted with contaminants of both natural and artificial origin. It is well known that precipitation is the main mechanism for removing contaminant. In studies of precipitation scavenging*, it is sometimes convenient to distinguish between rainout (in-cloud) and washout (below-cloud). Rainout comprises all processes within the cloud, and washout, the processes of removal of particles (mainly) by the rain below the clouds (Junge, 1963). It is recognized widely that below-cloud scavenging, although complex, is somewhat simpler than its in-cloud counterpart. The in-cloud scavenging which contributes most for cleansing the atmosphere of contaminant has not been treated adequately. The most used treatment of in-cloud scavenging until now has been by integral estimation of the removal of contaminant by cloud or rain elements. For example, Engelmann (1968) has set forth an in-cloud scavenging model, based on the assumption that the process by which contaminant is removed, can be

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* The general term scavenging will be used to denote the removal of particles from the atmosphere by precipitation processes, without specification of a particular physical mechanism (Gatz, 1966).

expressed by a simple exponential function with a removal rate constant* Ω. Thus the concentration of contaminant C remaining in the cloud is expressed by the equation:

$$C = C_o e^{-\Omega t}$$

where C_o is the contaminant concentration at time t=0. In this treatment, the individual processes of particle attachment to cloud and/or precipitations are not evaluated, and no input to the cloud is provided for after its initial formation.

1.1 PARTICLE ATTACHMENT PROCESSES

In considering the removal of contaminant from the atmosphere by rain, it is necessary to evaluate the mechanisms by which particles and gases may become attached to cloud or rain elements. Different microphysical mechanisms, Brownian diffusion, impaction, thermophoresis, electrical effects, turbulent diffusion, nucleation, may operate or predominate with certain particle characteristics, depending upon the cloud conditions.

Greenfield (1957) presented a theoretical model which indicates that particles with diameter larger than 2 μm are captured by raindrops under the influence of direct impact

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*The term removal rate constant (sec^{-1}) will be used to denote the removal of particles and/or contaminated cloud droplets from the cloud to the ground by precipiation elements.

collection, whereas smaller particles fail to collide with the raindrops, and so have to become attached to cloud droplets by diffusion processes. Diffusive attachment becomes fairly considerable in the case of Aitken particles ($r < 0.05$ μm) and ineffective for particles larger than about 0.05 μm radius. Slinn and Hales (1971) suggest that thermophoresis enhances the below-cloud rain scavenging of particles with radii between 0.01 and 1.0 μ. They also propose that electrical effects might even be more significant. Several writers (Sartor, 1967; Kamra, 1970, etc.) have developed solutions for discrete particles and drop charges, but their information appears insufficient to extrapolate their results to other charge values. In addition, there is a dearth of information on the charges and polarities of rain or cloud elements in different types of cloud (Gunn, 1952; Webb and Gunn, 1955; Krasnogorskaya, 1960, etc.). Therefore, the evaluations of electric effects remain somewhat uncertain.

Within the cloud, particularly under convective conditions, the process of turbulent diffusion may play an essential role. Authors (Saffman and Turner, 1956; Mazin and Ivanovskii, 1960; Levich, 1962) have suggested that the turbulent attachment rate of particles varies directly with the square root of the rate of energy dissipation. Ackerman (1968) shows the energy dissipation rates to be smallest for stratiform clouds and to be largest for convective clouds. Because the problem relates specifically to the dynamics of small droplets and particles, which must respond to a limited frequency band, it appears that the relevant energy dissipation may need more careful definition. Obviously, the mathematical and physical formulation of the problem of collision probabilities among particles in a turbulent medium is still to be done.

That nucleation is of prime importance in the removal of atmospheric contaminants is supported by many authors (Junge, 1963; Dingle, 1966; Hicks, 1966; Storebø, 1966, etc.) However the treatment of this physical process, from the standpoint of its function as a scavenging mechanism, is less satisfactory. Only Storebø and Dingle's (1973) work appears to be appropriate for predicting the nucleation scavenging of contaminants. The nucleation of droplets depends on i) aerosol characteristics (solubility, wettability, size, etc.), ii) atmospheric properties (supersaturation, temperature, etc.), and iii) air motions. In order to have a better understanding of the contribution of nucleation to precipitation scavenging, these parameters must be considered.

In order to obtain some information on qualitative prediction of scavenging, many empirical models have been proposed.

1.2 CONCENTRATION - TOTAL PRECIPITATION AMOUNT RELATIONSHIPS

Table 1 (Makhon'ko, 1966) summarizes proposed formulae relating the radioactivity concentration in cloud water to the total amount of precipitation.

The parameters in the formulae were selected empirically in each individual case. The scavenging capacity of rain originating from different types of cloud and microphysical mechanisms involved in the scavenging process were not examined. Therefore, these proposed formulations should be viewed only as extremely rough indications of the precipitation scavenging phenomena.

In Table 1, C'_o and C' denote the radioactivity concentration in cloud water before and after the onset of rain, respectively, and h is the total amount of precipitation received on the day in question. The parameters a', b', C'_1 and ζ in the formulae are determined from observational data. F and λ' denote respectively the rate of formation and the decay constant for Be^7. H' is the height of the origin of the rain, m is the radioactivity of an individual particle, n' is the concentration of particles, R is the raindrop radius, E' is the collision efficiency, P is the amount of fallout, and q is the radioactivity concentration in air.

Table 1

Relationship between concentration of radioactivity and diurnal amount of precipitation (after Makhon'ko, 1966)

Formula No.	Formula	Reference
1	$C' = \frac{C'_o}{h}(1-e^{-a'h})$	Damon and Kuroda, 1954
2	$C' = \frac{F}{\lambda+a'h}$	Gustafson, et al., 1961
3	$C' = q\frac{a'}{h^{b'}}$	Perison, et al., 1960
4	$C' = a'-b'h$	Szalay and Berenyi, 1958
5	$C' = \frac{C'_o}{h}(1+a'h-e^{-b'h})$	Miyake, et al., 1960
6	$C' = \frac{C'_o}{h}(1-e^{-a'h})$	Miyake, et al., 1957
7	$C' = C'_o e^{-a'h}$	Yano, 1961
8	$C' = \frac{H'mn'}{h}[1-\exp(-\frac{3}{4}\frac{E'}{R}h)]$	Yano and Naruse, 1956
9	$C' = q[\frac{a'}{h} + \frac{b'}{h}(1-e^{-\zeta h^{1.2}})]$	Moeken and Alderhorst, 1963
10	$C' = q(\frac{a'}{h} + b'h^{0.2}-\zeta h^{1.4})$	Moeken and Alderhorst, 1963

Table 1 (Cont.)

Formula No.	Formula	Reference
11	$C' = C'_o + \frac{1}{h}[\frac{C'_1}{b'}(1-e^{-b'h})$	
	$+ qH(1-e^{-b'h})]$	Makhon'ko, 1964
12	$P = a'q^{b'}h^{\zeta}$	Storebø, 1957
13	$P = q(a' + b'h)$	Storebø, 1957

These early works fail to provide any information on the relationship between temporal variations of contaminant concentration and rainfall rate, particularly in convective rains. Since both rainfall intensity and contaminant concentration in rain samples are products of the precipitation generation system, their relationship could be a key to understanding the precipitation scavenging and the structure of the convective clouds.

1.3 CONTAMINANT CONCENTRATION VARIATIONS DURING CONVECTIVE RAINS

Dingle and Gatz collected rain samples and measured rainfall intensities during convective rains in a series of field experiments from 1964 to 1968. Their observations show that the contaminant concentrations in rain water samples collected sequentially at a station are most frequently correlated negatively with rainfall intensities. This is the inverse relationship to which we shall refer. But high concentration associated with high rainfall intensities have also been observed from time to time. This is the "direct relationship". Field observations from other researchers reveal similar results (Kruger and Hosler, 1963; Huff and Stout, 1964; Bleeker, 1966, etc.). Explanations of the observations vary among the above researchers, some of whom require that different scavenging mechanisms explain the different kinds of relationship.

Following Gatz and Dinge (1971), it is proposed here that both direct and inverse relationships can be expected as a result of rain production and scavenging processes interacting with the circulation dynamics of convective systems. Two broad basic classes of consideration enter: (1) the processes of cloud drop nucleation and growth interacting with and involving scavenging processes, and (2) the distributive effects of convective circulation patterns interacting with the raindrop fall and coalescence processes. The cloud microphysics and scavenging processes generally lead to the prediction of an inverse relationship between concentration of contaminant and rainfall rate. For example: (a) nucleation of cloud droplets; the most active nuclei (generally, though not necessarily only the largest particles) form large droplets early in the lower part of the cloud, and precipitate out carrying relatively little water (low rainfall rate) and relatively high contaminant concentration; smaller particles nucleat later and circulate higher; (b) condensation and evaporation; condensation contributes pure water (increased rainfall rate) to the growing droplets, and evaporation does the reverse, each with respect to a constant mass of contaminant (the initial nucleus of each droplet), hence these processes both tend to produce a negative correlation between rainfall rate and concentration.

At the same time, the convective circulation tends to produce a maximum concentration of contaminant in the core of the major updrafts, diluting this outward by turbulent diffusion and lateral entrainment of relatively clean air.

The result is a variation of contaminant concentrations in the horizontal direction. This distribution also shifts in the vertical direction, and falling drops may vary their contaminant concentrations as they grow by coalescence along their fall paths.

The literature review given above indicates that the previous research on this subject has scarcely passed the qualitative stage. Therefore it is evident that quantitative evidence may contribute to further understanding of the causes for both types of relationship.

Convective clouds are different from stratiform clouds in many important aspects such as cloud water content, vertical air velocity, cloud structure, circulation patterns and life cycle, etc. To develop an in-cloud scavenging model particularly for convective clouds is an extremely complex work by virtue of its many interacting and competing mechanisms. Therefore the few simplified models developed so far are only applicable to stratiform clouds. Following is a review of these models.

1.4 IN-CLOUD SCAVENGING MODEL

1.4.1 Makhon'ko's model (1967)

This model is based on a differential equation describing the variation in concentration of radioactive particles with time resulting 1) from the attachment of particles to cloud droplets and 2) from the divergence of contaminant mass flow.

The result is

$$C' = \psi_o + \psi_1 e^{-\Omega_o t} + \psi_2 e^{-\Omega_1 t}$$

where C' is the contaminant concentration in precipitation, ψ_o, ψ_1 and ψ_2 are parameters depending on the air vertical velocity, cloud thickness, cloud droplet concentration, and rainfall intensity, t is the precipitation duration. Ω_o and Ω_1 are parameters characterizing the removal rate constant in and below cloud regions, respectively. This expression shows that after rain begins, the contaminant concentration in precipitation decreases because of its removal by the rain and tends to a constant limit, ψ_o, which is due to the establishment of equilibrium: the amount of contaminant removed is equal to that taken in by the inflow of air.

In his model, the following assumptions are made:

1) the cloud water remains constant and consists of monodispersed droplets with a number density of $300/cm^{-3}$.
2) contaminant particles are also monodispersed.
3) particles attached to cloud droplets are immediately removed from the cloud to the ground.
4) parameters Ω_o and Ω_1 are empirically determined.

1.4.2 Davis model (1972)

This model suggested by Davis is based on 1) the consideration of the production and decay of cosmogenic radionuclides, 2) its transfer to cloud water, and 3) its removal by snow or rain. The results are

$$\text{i)} \quad N'_a = \underbrace{\frac{\theta_1 N'_o}{\theta_o+\theta_1} e^{-(\theta_o+\theta_1)t}}_{A} + \underbrace{\frac{P_r}{\theta_o+\theta_1}}_{B}$$

where N'_a is the air concentration, θ_1 is the attachment rate constant,* θ_o is the decay rate constant, N'_o is the equilibrium concentration when decay is equal to production, t is the precipitation time, and P_r is the rate of production. This expression indicates that the particle concentration in cloud air decreases because of its attachment to cloud droplets and because of its decay (term A), and increases because of the production of nuclides (term B).

$$\text{ii)} \quad N'_w = \underbrace{N'_{wo} e^{-(\theta_2+\theta_o)t}}_{A} + \frac{\theta_1^2 N'_o e^{-(\theta_o+\theta_1)t_o}}{(\theta_o+\theta_1) \quad (\theta_2+\theta_1)} e^{-\theta_o t} .$$

$$\{\underbrace{e^{-\theta_1 t}}_{B} - \underbrace{e^{-\theta_2 t}}_{C}\} + \frac{\theta_1 P_r}{(\theta_o+\theta_1)(\theta_2+\theta_o)} (1-\underbrace{e^{-(\theta_o+\theta_2)t}}_{D})$$

where N'_w is the cloud water concentration of nuclides, N'_{wo} is the water concentration at time t_o, t_o is the in-cloud time before precipitation begins, and θ_2 is the removal rate constant. This expression is composed of four terms. Term A indicates that N'_w varies because of the decay of particles and because of its removal by rain. Term B indicates that

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*The term attachment rate constant (sec^{-1}) will be used to denote the attachment of particles to cloud droplets. When particles become attached to raindrops, the attachment rate constant is equivalent to removal rate constant.

N'_w varies because of particles attached to cloud droplets. Term C indicates that N'_w varies because of its removal by rain. Fainally, term D indicates that N'_w varies because of the production of particles.

iii) $\frac{dN_g}{dt} = \theta_2 N'_w \, e^{-\theta_o t}$

where $\frac{dN_g}{dt}$ denotes the rate of deposition on the ground. The assumptions made in this model are very similar to those made in Makhon'ko's model. A significant difference between Davis' model and the models by Engelmann and by Makhon'ko' is that Davis' distinguishes between the attachment rate constant and the removal rate constant. At this point, it is appropriate to point out the physical reasoning why there is a need for this distinction. As is well known, cloud droplets are characterized by small size, thus they have a negligible terminal fallspeed. Therefore particles attached to cloud droplets would remain in the cloud and could not be immediately removed from the cloud to the ground.

In view of the existing models, it is apparent that the presenting available conceptual framework for in-cloud scavenging is inadequate. A number of disadvantages can be noted: 1) the assumption of uniform cloud droplets and particle sizes, 2) the size-time-independent attachment rate constant and the size-time-independent removal rate constant, 3) the lack of distinction between the attachment rate constant and the removal rate constant, and 4) the integral estimation of contaminant removed by precipitation using unrealistic parameterization of the microphysical processes.

1.5 PURPOSE OF THE RESEARCH

The review of in-cloud scavenging models indicates a need for additional study. Previously it has been assumed that in-cloud scavenging was a single-step process (Makhon'ko, 1967; Engelmann, 1968). However, not all the microphysical processes are treated adequately in this way. In this study, the concept and the theoretical approach are introduced to show that the in-cloud scavenging should be viewed as a multi-step process. Based on this concept, a numerical model for stratiform clouds is proposed.

As mentioned in the previous section, most writers on the subject of direct and inverse relationships appear to have assumed that the two types of relationships are produced by different mechanisms. To provide quantitative evidence regarding the mechanisms from which these two types of relationships can result, a numerical model is developed. This model is intended to provide information for a better understanding of the relationships between the contaminant concentration and the rainfall intensity and of the resulting deposition patterns.

1.6 THE APPROACH TO THE PROBLEM

The in-cloud precipitation scavenging processes that we would like to discuss are

1) the depletion of contaminants in cloud air through various microphysical mechanisms: Brownian motion, turbulent coagulation, and impaction. These processes determine the rates at which the particles became attached to cloud and/or rain elements. Since all of the rate constants are size-dependent, a realistic approach requires consideration of the size spectra of drops (cloud and rain) and particles.
2) the removal of contaminanted cloud droplets and contaminant by an independently generated overriding rain system.

The rate constants of the attachment and removal processes are treated as time-dependent in the computations. The analytical expressions are presented in Chapter 2.

In Chapter 3 a model of a steady-state convective cloud is presented. As noted above, this is a complex problem. To reduce it to a manageable form parameterization is necessary. In the present instance, the results from the stratiform cloud model aid in the parameterization of the microphysics. As a result of these simplifications it is possible to construct a plausible model in which the processes of raindrop fall, coalescence, and breakup can be considered, and a spacial distribution of contaminant in the cloud droplets can be assumed. Although this particular model is undoubtedly much simpler than a real convective cloud, the assumptions permit the model to fulfill the requirement that it produces both direct and indirect relationships between the rainfall rate and the contaminant concentration in the rain water.

CHAPTER 2

A MODEL FOR RAIN SCAVENGING FROM STRATIFORM CLOUDS

It is recognized that in-cloud scavenging is more important for cleansing the atmosphere of contaminant than is below-cloud scavenging. For the below-cloud case, the theoretical predictions are available. (Zimin, 1964; Engelmann, 1968; Slinn and Hales, 1971). The situation for the in-cloud case is less satisfactory. Beginning with the paper by Greenfield (1957), a number of papers have appeared which treat various parts of the problem (e.g., Goldsmith, et al., 1963; Hicks, 1966; Vittori and Prodi, 1967; Slinn and Hales, 1971; Davis, 1972), but no comprehensive treatment has so far appeared. The purpose of the present study is to put forward a simplified model of in-cloud scavenging which is primarily based on continuity equations, incorporates the diffusive attachment, impaction and accretion processes and includes consideration of particle and cloud droplet size spectra. Owing to the lack of quantitative information on phoretic and electrical influences on the attachment rate, the contribution to the total in-cloud scavenging from these processes are ignored.

2.1 THE MODEL

In the present model, the cloud is envisaged as an assembly of droplets intermingled with a particulate contaminant some of which is free-floating in the cloud air and some of which is attached to the droplets. Overriding rain, independently generated, removes contaminant particles of both classes.

The temporal variation of the contaminant concentration in each category is expressed by means of an ordinary differential equation:

$$\frac{dNa_i}{dt} = - \left[\sum_{j=1}^{M} \alpha_{ij} + \sum_{k=1}^{N} (\beta_{ik} + \gamma_{ik}) \right] Na_i \qquad \text{for each } i \qquad (1)$$

$$\frac{dNc_{ij}}{dt} = \alpha_{ij} Na_i - Nc_{ij} \sum_{k=1}^{N} \lambda_{jk} \qquad \text{for each } i \text{ and } j \qquad (2)$$

$$\frac{dNr_{ik}}{dt} = \sum_{j=1}^{N} \lambda_{jk} Nc_{ij} + (\beta_{ik} + \gamma_{ik})\, Na_i \qquad \text{for each } i \text{ and } k \qquad (3)$$

where

Na = number density of contaminant particles in the cloud air, cm^{-3}

Nc = number density of contaminant particles attached to cloud droplets, cm^{-3}

Nr = number density of contaminant particles removed by raindrops, cm^{-3}

α = diffusive attachment rate between particles and droplets (sec^{-1})

β = diffusive attachment rate between particles and raindrops, sec^{-1}

γ = impact collection rate for particles by raindrops, sec^{-1}

λ = rate of accretion of droplets by raindrops, sec^{-1}

i = particle size class index (i = 1...30)

j = cloud droplet size class index (j = 1 to M = 17)

k = raindrop size class index (k = 1 to N = 17)

and the system conserves contaminant mass.

This system of equations would have the attractive features of 1) giving greater accuracy in the calculations and 2) providing an inventory of particle sizes. At the same time, a large computing time must also be expected. In order to speed the calculations for a large set of equations, we seek some approximations which will both reduce computing time and maintain accuracy within a certain limit. To this end, a number of approximations to (2) and (3) will be made.

The first term on the right-hand side of (2) is rewritten as

$$Na_i \sum_{j=1}^{M} \alpha_{ij}$$

which computes the total number of particles of each ith class interacting with the entire cloud droplet population. To simplify the calculation of the accretion term, we then define the weighted mean accretion rate, $\bar{\lambda}$, by the following formula:

$$\bar{\lambda} = \frac{\sum_{j=1}^{M} Nc_j \left(\sum_{k=1}^{N} \lambda_{jk} \right)}{\sum_{j=1}^{M} Nc_j}$$

As the results show, the weighted mean value of the accretion rate approximately corresponds to $\sum_{k=1}^{N} \lambda_{jk}$ computed for the mean cloud droplet radius in the assumed size spectrum. As described later, (see Section 3), the assumed size spectra have such a distribution that the number density of cloud droplets and raindrops is larger the smaller the drops. Thus, the use of $\bar{\lambda}$ is expected to influence the accuracy of the computations only slightly.

As a consequence of these simplifications, the basic continuity equations (1), (2), and (3) can be rewritten as:

$$\frac{dNa_i}{dt} = - \left[\sum_{j=1}^{M} \alpha_{ij} + \sum_{k=1}^{N} (\beta_{ik} + \gamma_{ik}) \right] Na_i \quad (4)$$

$$\frac{dNc_i}{dt} = Na_i \sum_{j=1}^{M} \alpha_{ij} - \bar{\lambda} Nc_i \quad (5)$$

$$\frac{dNr_i}{dt} = \bar{\lambda} Nc_i + \left[\sum_{k=1}^{N} (\beta_{ik} + \gamma_{ik}) \right] Na_i \quad (6)$$

A physical interpretation of the simplified equations (5) and (6) is that the cloud droplets and the raindrops work as two individual ensembles and interact with each other at a single rate $\bar{\lambda}$ defined by the equation. In doing so, the individual interactions between raindrops and cloud droplets could not be distinguished, and the number of particles of each size removed by kth class raindrops and attached to jth class cloud droplets could not be specifically inventoried.

If the approximations discussed above are acceptable, then the calculations are greatly simplified, and the number of particles remaining in cloud air, attached to cloud droplets and removed by raindrops remains to be identified.

2.2 ANALYSIS

In the course of our analysis, we propose to solve the problem analytically. Obviously, as long as α, β, γ, and $\bar{\lambda}$, (hereafter the indices are omitted for compactness; it must be remembered that the coefficients are evaluated over the entire spectra) are considered constants in a single time interval, the equations (4), (5) and (6) can be integrated in time. If the contaminant is allowed to interact with the cloud for a time, t_c, before precipitation begins, then the air concentration can be written directly:

$$Na(t_c) = Na\ (0)e^{-\alpha t_c} \qquad (7)$$

where Na(0) is the initial particle concentration in the cloud air. The number of particles associated with the droplets prior to the rain is

$$Nc(t_c) = Nc(0) + Na(0)\ (1-e^{-\alpha t_c}) \qquad (8)$$

where Nc(0) is the number density of the particles that serve as condensation nuclei as the cloud is formed.

After the onset of rain, the number density of contaminant particles remaining in the cloud air at time t, measured from the onset of rain, becomes

$$Na(t) = Na(0)\ e^{-\alpha t_c}\ e^{-(\alpha + \beta + \gamma)t} \qquad (9)$$

The number density of contaminant particles associated with the remaining cloud droplets is then

$$Nc(t) = [Nc(0) + Na(0)\ (1-e^{-\alpha t_c})]e^{-\bar{\lambda}t} +$$

$$\frac{\alpha Na(0)e^{-\alpha t_c}}{\bar{\lambda} - (\alpha+\beta+\gamma)}\ (e^{-(\alpha+\beta+\gamma)t} - e^{\bar{\lambda}t}) \qquad (10)$$

and the number of pollution particles per unit volume removed by raindrops can be written as

$$Nr(t) = [Nc(0) + Na(0)]\ (1-e^{-\bar{\lambda}t}) +$$

$$\frac{Na(0)e^{-\alpha t_c}}{\bar{\lambda} - (\alpha+\beta+\gamma)}\ [e^{-\bar{\lambda}t} - e^{-(\alpha+\beta+\gamma)t}]\ [\bar{\lambda} - (\beta + \gamma)] \qquad (11)$$

Equations (9), (10), and (11) are evaluated over the entire particle size spectrum.

Evaluation of Parameters.

The rate constants must be evaluated in terms of the respective physical processes.

a) Diffusive attachment. The particles become attached to the droplets by Brownian and turbulent diffusion, each of which contributes to the value of α. If α_B is the rate of attachment contributed by Brownian diffusion, then

$$\alpha_B = \Sigma \frac{kT}{3\eta} \left(\frac{1 + a\ell/r_p}{r_p} + \frac{1 + a\ell/r}{r} \right) (r_p + r) \, M_c$$

(Byers, 1965) for each i and j (12)

where

r_p and r are particle and droplet radii of each size class, respectively;

a is the Cunningham correction factor (=0.9);

ℓ is the mean free path of air molecules;

M_c is the number density of cloud droplets of each size class, cm^{-3};

k is the Boltzmann's constant;

T is the absolute temperature; and

η is the air viscosity.

Levich (1962), in the investigation of the effect of air turbulence on the coagulation of particles, pointed out that the attachment of particles is enhanced under the influence of small eddies. In the present study, we adopt the Levich formulation:

$$\alpha_T = \Sigma 14.1\ (r_p + r)^3\ (\frac{\varepsilon}{\nu})^{1/2}\ M_c \qquad \text{for each } i \text{ and } j \qquad (13)$$

for the contribution, α_T, of air turbulence to the attachment rate. Here ε is the rate of energy dissipation, ν is the kinematic viscosity. The quantity ε is dependent upon the nature of the cloud considered. Ackerman (1968) gives a value, ε, (for the inertial subrange) of 6.4 $cm^2\ sec^{-3}$ as the average for stratiform cloud. The assumption of inertial subrange turbulence may not be valid for several reasons, one of which is the frequency amplitude sensitivity of particles, i.e. resonance. We use it lacking a better estimate, but we do so with reservations. We also consider the turbulent and Brownian contributions to be additive, hence

$$\alpha = \alpha_B + \alpha_T \qquad (14)$$

To complete the evaluation of the diffusive attachment rate, α, the size spectra of the contaminant particles and the cloud droplets are required. For the latter Khrgian and Mazin (1961) have proposed,

$$n(r) = ar^2e^{-br} \qquad (15)$$

Here $n(r)$ is the number of droplets per unit volume in the radius interval between r and $r + dr$, and

$$a = (1.45\ \bar{r}^{-6})\ Q/\rho_\ell$$

$$b = 3\bar{r}^{-1}$$

Q is the liquid water content, gm cm^{-3}

ρ_ℓ is the water density and

$\bar{r} = \frac{1}{N} \int_0^\infty r\, n\, (r)\, dr$ is the average radius.

The particle size spectrum is chosen to be log-normal, and is specified in terms of the geometric modal radius, r_m, and the geometric standard deviation, σ ($\sigma = 2.5$). For the present model we have set $r_m = 10^{-5}$ cm following Junge (1963). This is a parameter of the model; if it is displaced to smaller values, then enhancement of diffusive attachment is expected, and conversely, if r_m is larger, the diffusive mechanism must be less effective. Therefore

$$N_p = A \exp \left[-0.5 \left(\ln \frac{r_p}{r_m}\right)^2 (\ln \sigma)^{-2}\right] \qquad (16)$$

where N_p is the number of particles per unit volume in the radius interval between r_p and $r_p + dr_p$. For the computations, the size spectrum is composed of 30 classes at intervals of

$r_{n+1} = 2^{1/3}\, r_n$, and r_0 is chosen at 10^{-6} cm.

The same treatment is used to evaluate the diffusive attachment between particles and raindrops.

(b) Raindrop size distribution. Marshall and Palmer (1948, hereafter referred to as the M-P expression) first demonstrated that the average number size distribution of stratiform rain may best be expressed by the exponential function

$$n(x) = n_0 e^{-\Lambda x}$$

where $n(x)$ is the number concentration per unit volume per unit size interval of raindrop diameter x, and

$$n_0 = 0.08 \text{ cm}^{-4}, \quad \Lambda = 41 I^{-0.21} \text{ cm}^{-1}.$$

where I is the rainfall rate in mm/hr.

Recent investigations by Sekhon and Srivastava (1970) of doppler radar observations in thunderstorm rainfall also suggest that the raindrop size distribution is exponential in nature and may be represented by M-P expression but with $n_0 = 0.07\ I^{0.37}$ cm^{-4} and $\Lambda = 38\ I^{-0.14}$ cm^{-1}.

An increase in stratiform rainfall rate is accompanied solely by a decrease in the slope (Λ) on a plot of $\ln n(x)$ vs. x, thus implying increased concentrations of larger drops. A corresponding increase in rate of thunderstorm rainfall is attributable to both an increase in the intercept, $\ln n_0$ on such a plot, and a somewhat smaller decrease in the slope. In the computations presented in this study, we take $n_0 = 0.08$ cm^{-4}, and $\Lambda = 41\ I^{-0.21}$ cm^{-1}.

c) Impact collection. The impact collection rate is

$$\gamma = \Sigma[\pi R^2\ E(r,R)\ (V_R - V_r)\ N_R\ \Delta R] \qquad \text{for each } i \text{ and } k \qquad (17)$$

where V_R is the terminal velocity of raindrops of radius R (Dingle and Lee, 1972); V_r is the terminal velocity of particles of radius r; E(R,r) represents the collision efficiency; and $N_R\ \Delta R$ is the number of raindrops per unit volume in the size range R to R + ΔR. Values of E (R, r) are derived from the table compiled by Mason (1971).

d) Accretion. The removal of cloud droplets by rain is given by equation (17) when R denotes the raindrop radius, r the cloud droplet radius, and when λ is substituted for γ.

2.3 RESULTS

One of our basic assumptions is the conservation of mass. We must ensure that the mass is indeed conserved as computations progress. The number of particles Na(t), Nc(t) and Nr(t) of each class is computed in each time step, whereby the mass can be determined. The relation $\Sigma Na(t)+Nc(t)+Nr(t)=Na(0)$ must hold for any time step and any size class. Since at the time t=0, we initiate our computation, the total mass is fixed, thus the number of particles of each class, Na(0), is also prescribed by the assumed distribution.

Four cases have been selected to bring out the scavenging effects of various parameters of the model. In the computations, the cloud droplet spectrum and size distribution of pollution are assumed to be time-independent for a single time step, and the term Nc(0) of Equations (8), (10), and (11) is not accounted for. To simplify our computations, we assume that the parameters α, β, γ, $\bar{\lambda}$ remain constant for a short time interval and are adjusted for each time step. The time step is mainly determined by the amount of water removed from the cloud. In our computations, it is shown that the weighted accretion rate, $\bar{\lambda}$, ranges from 10^{-4} (sec^{-1}) to 10^{-3} (sec^{-1}). For rainfall rates less than 4 mm/hr, the cloud water removed every two minutes does not significantly change the cloud droplet concentration. Therefore, the

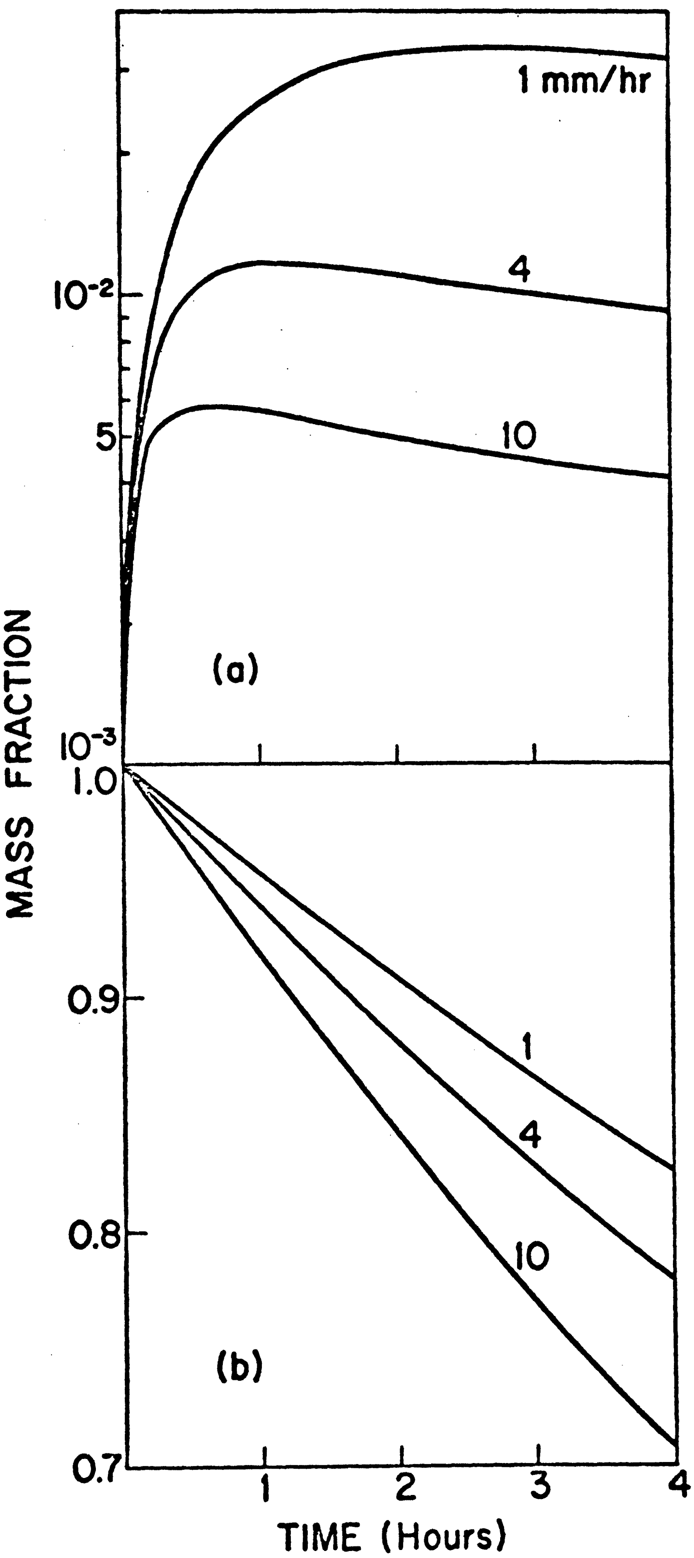

Fig. 1. Case I. Cloud water constant (replenished): (a) fraction of mass attached to cloud droplets and (b) fraction of mass remaining in cloud air as a function of time.

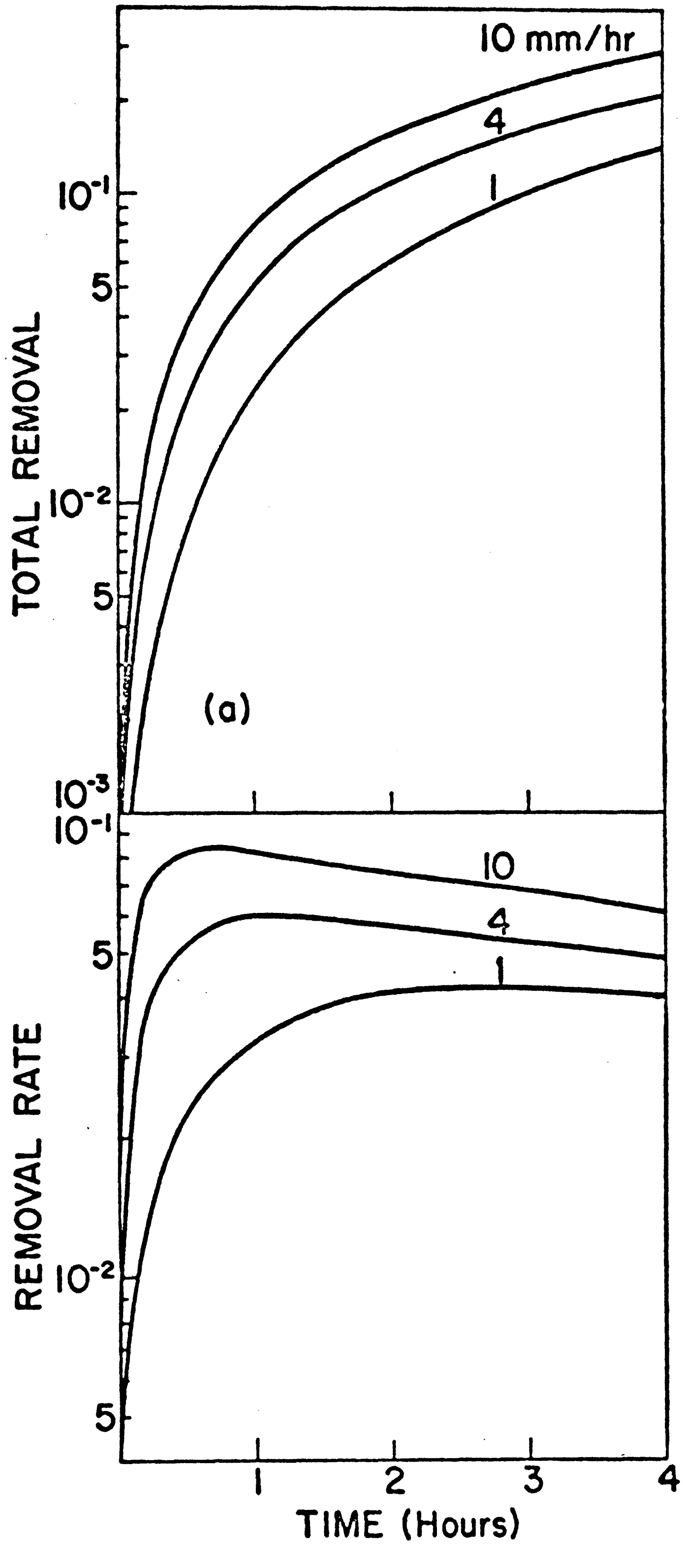

Fig. 2. Case I. Cloud water constant (replenished): (a) integral fraction of mass removed by rainfall and (b) removal rate (mass fraction per hour) as a function of time.

parameters may be considered constant for this time interval. For rainfall rates greater than 4 mm/hr, the time step is reduced to one minute and the parameters are readjusted accordingly.

a) Case I: Steady state cloud: cloud water replenished at rate exactly equal to rate of removal of cloud water by falling rain. Removal of pollution particles by a) Brownian and turbulent attachment to cloud droplets and falling rain, and b) removal of cloud water with attached pollution by falling rain. Results for this case are shown in Fig. 1 and Fig. 2. The fraction of pollution mass remaining in the cloud region after time t at various rainfall rates is given in Fig. 1b.

As it is found, the impact-collection of pollution particles by the falling raindrops is not an effective means of removing them from the air, since particles smaller than 2μm radius are not captured by raindrops, (see Appendix A) and particles greater than 2μ radius have a very low concentration. As shown in Fig. 1a, the amount of contaminant attached to cloud droplets is initially zero. It increases, reaches a maximum after a time which is dependent upon the relative magnitudes of attachment rate and scavenging rate, and then decreases gradually. Fig. 2a shows the trend of the fraction of integral mass removed from the cloud at various rainfall rates.

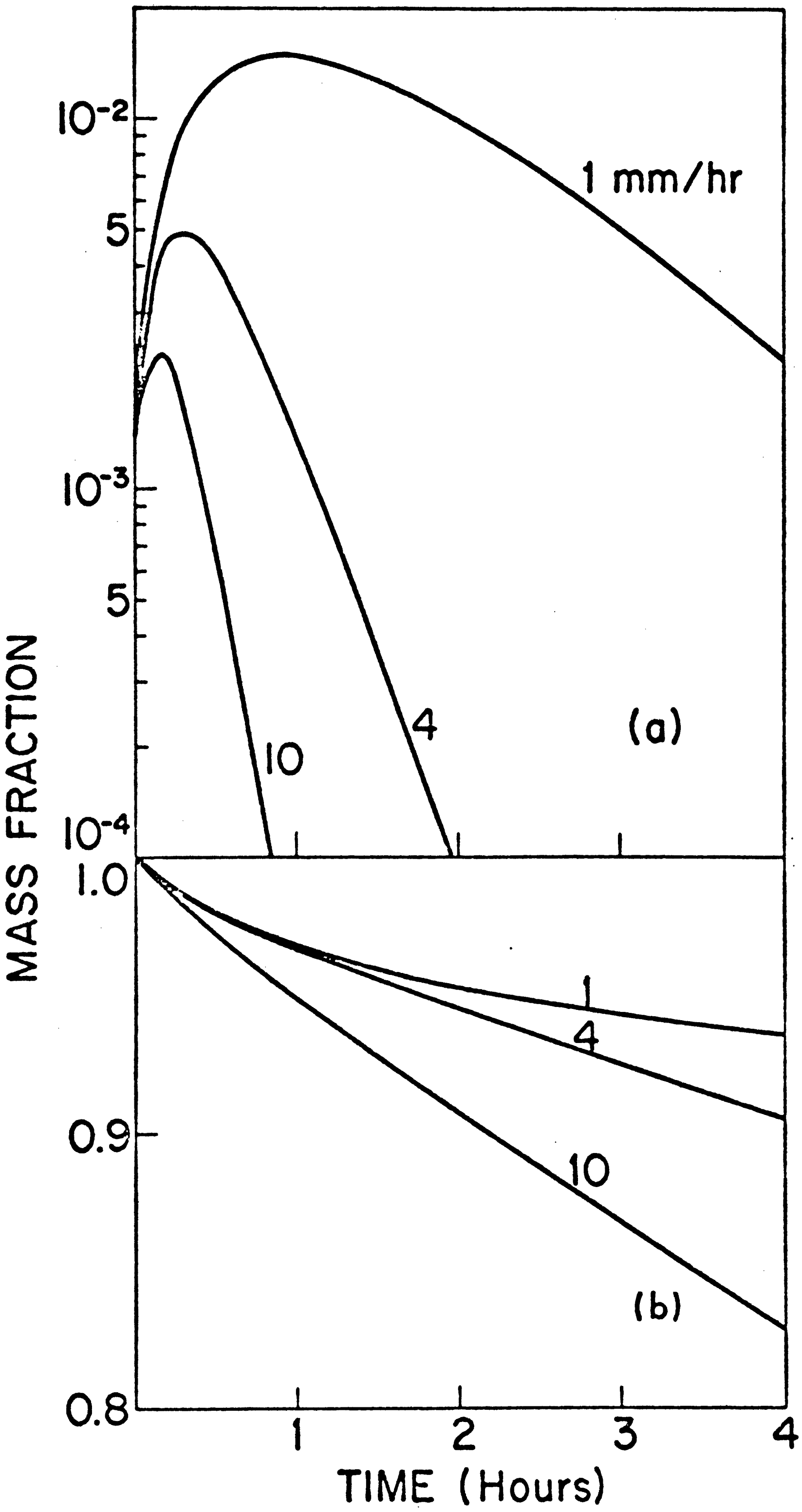

Fig. 3. Case II. Decreasing cloud water: (a) fraction of mass attached to cloud droplets and (b) fraction of mass remaining in cloud air as a function of time.

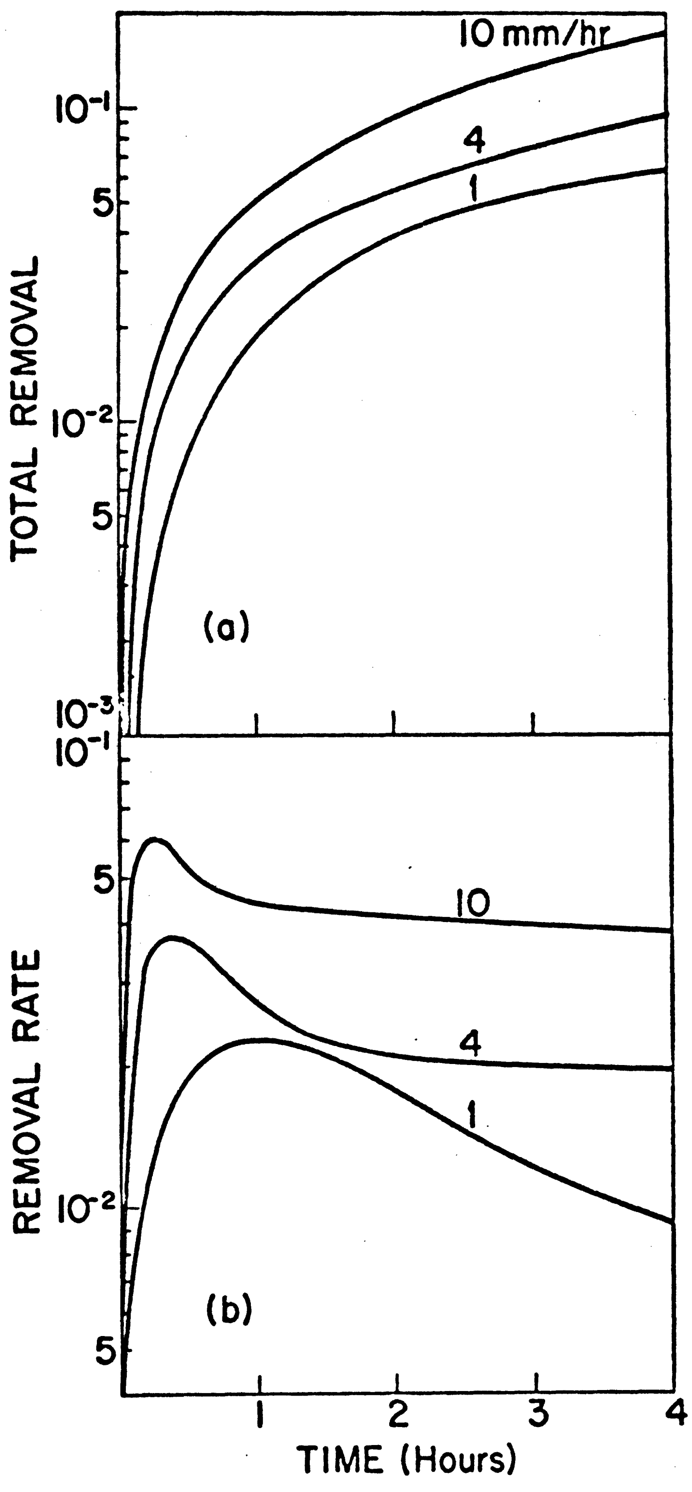

Fig. 4. Case II. Decreasing cloud water: (a) integral fraction of mass removed by rainfall and (b) removal rate (mass fraction per hour) as a function of time.

It is found that about 29% of the contaminant is scavenged by a rainfall of intensity 10 mm/hr in four hours. The removal rate (mass fraction per hour) is shown in Fig. 2b. The heavier the precipitation the earlier the maximum removal rate occurs.

b) Case II: Similar to Case I except that the cloud is not replenished. In this case, the concentration of cloud droplets decreases with time. This decrease in drop concentration in turn leads to a lower rate of attachment of particles to cloud droplets.

Figs. 3 and 4 show the results for Case II. The trend of pollution remaining in the air is similar to that in Fig. 1b. The major difference between Fig. 1b and Fig. 3b is the relatively small amount of material scavenged in the latter. The obvious reason for this is that cloud water is decreased. The maximum removal rates occur at an earlier stage in Case II than in Case I. Also the diffusive attachment processes lead to an earlier maximum of pollution mass acquired by cloud droplets in Case II than in Case I (Fig. 3a).

c) Case III & IV: Case III is similar to Case I except that the particulate contaminant has intermingled with cloud droplets for two hours prior to the onset of rain. Case IV is similar to Case III except that the cloud is non-steady as in Case II.

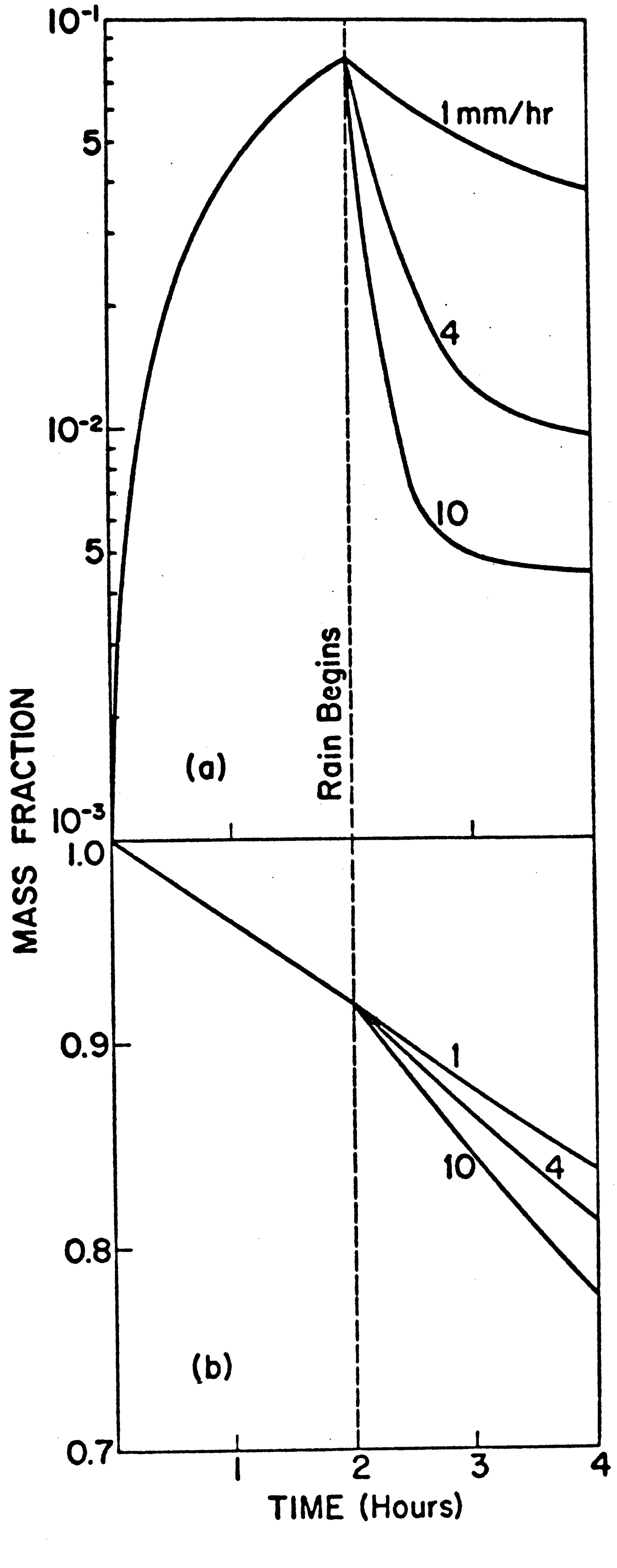

Fig. 5. Case III. Cloud water constant (replenished) with rain beginning at time t=2 hr: (a) fraction of mass attached to cloud droplets and (b) fraction of mass remaining in cloud air as a function of time.

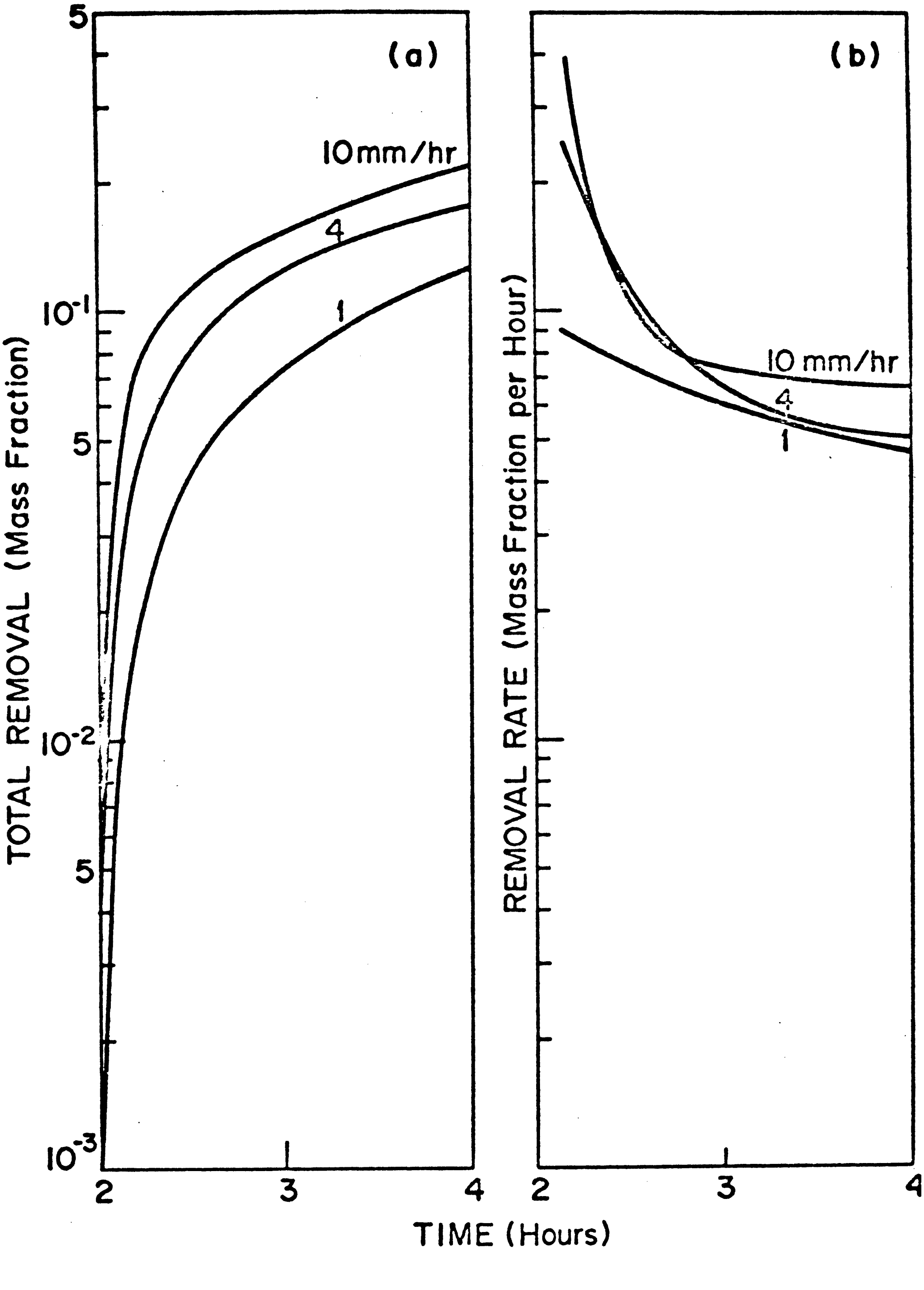

Fig. 6. Case III. Cloud water constant (replenished) with rain beginning at time t=2 hr (see Fig. 5): (a) integral fraction of mass removed by rainfall and (b) removal rate as a function of time.

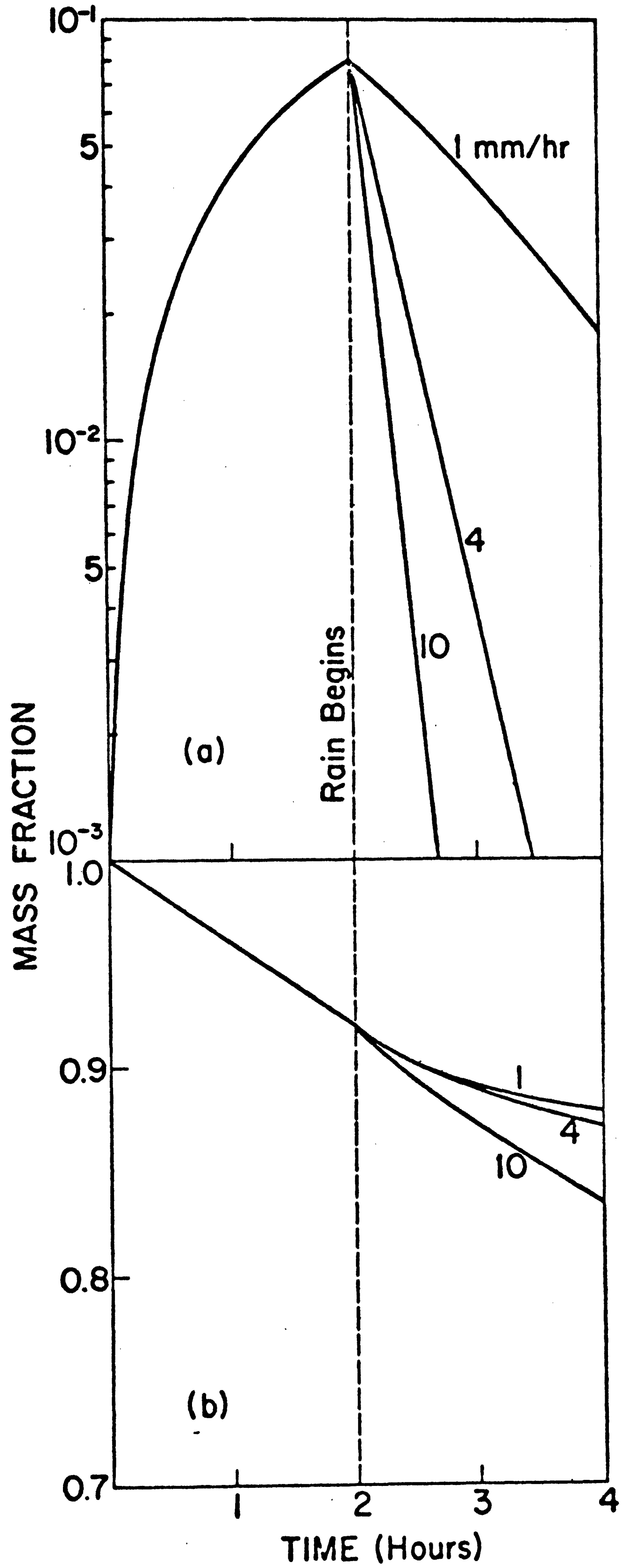

Fig. 7. Case IV. Decreasing cloud water with rain beginning at time t=2 hr: (a) fraction of mass attached to cloud droplets and (b) fraction of mass remaining in cloud air as a function of time.

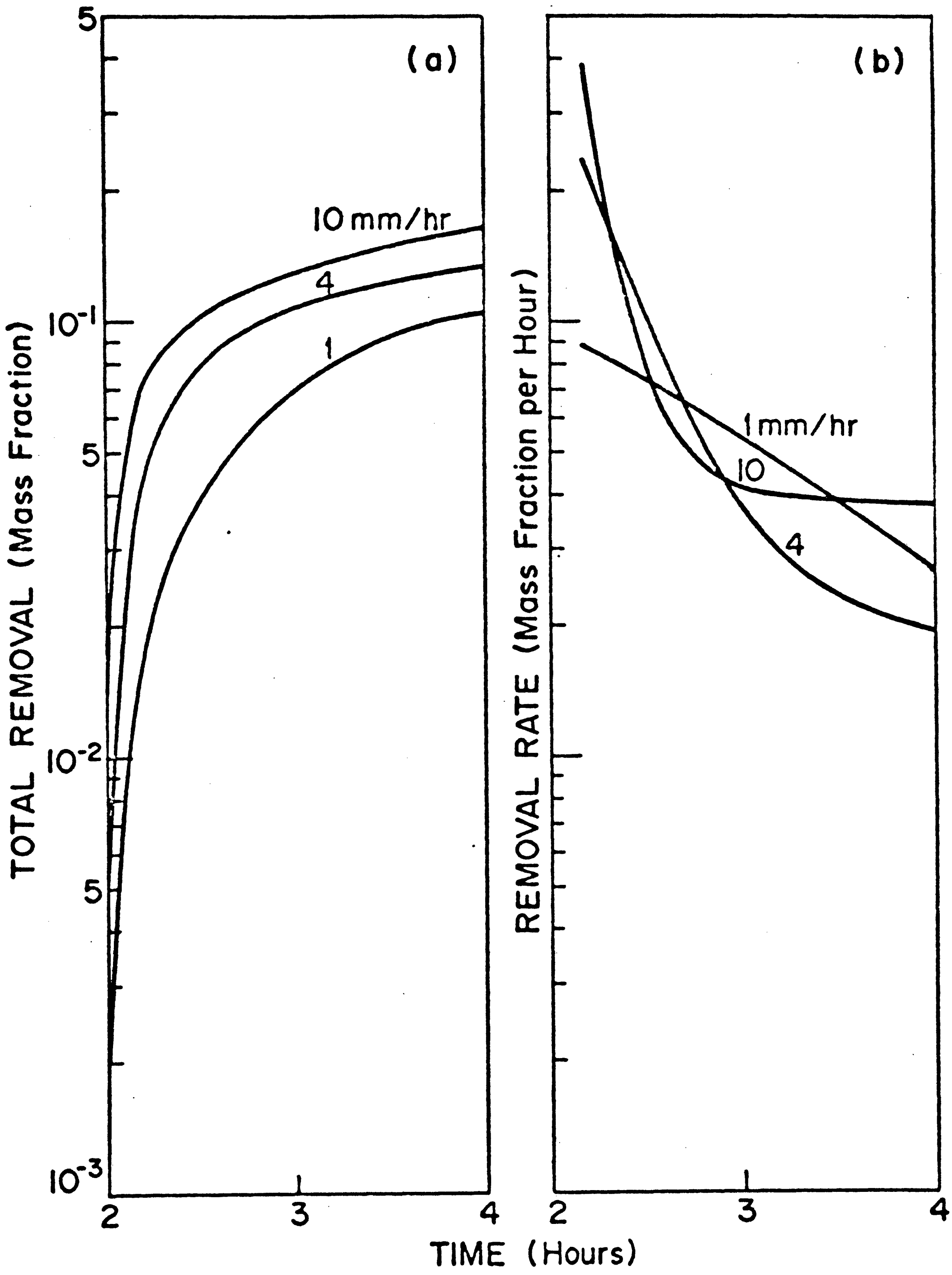

Fig. 8. Case IV. Decreasing cloud water with rain beginning at time t-2 hr (see Fig. 7): (a) integral fraction of mass removed by rainfall and (b) removal rate as a function of time.

In these cases, the in-cloud mixing time t_c is two hours, and rain falls through the polluted cloud for another two hours. Results for these cases are shown in Figs. 5, 6 and Figs. 7, 8. What is effectively demonstrated by these modeled cases is the increased removal of pollution mass where preliminary interaction is allowed. For example, in Case III (Fig. 5b), after two hours during which the rainfall rate is 10 mm/hr, the air contains 77% of initial pollution. This contrasts with Case I (Fig. 1b) where approximately 84% of initial pollution still resides in the air. One important difference brought out by Cases III and IV is that the contaminant mass fraction attached to cloud droplets has a completely different pattern from Case I or Case II. The same feature appears in the removal rate of pollution mass. This is due to the fact that by the time rain starts, the cloud droplets have already been acquiring contamination for two hours.

d) Rainout efficiencies. In considering the cleansing of the air by rain, noting the intimate involvement of the processes of cloud droplet nucleation, growth and the eventual production of rain from the contaminant-bearing air, it is recognized that the "scavenging" of water and of contaminant should be more or less proportionate. Junge (1963) introduces the equation

$$K = \chi_0 E \frac{\rho_\ell}{Q} \tag{18}$$

where K is the concentration of a specified contaminant in rain water, χ_0 is the concentration of the same contaminant originally in the air, and E is the rainout efficiency (Junge, 1963, Engelmann, 1968). No measurements of E are available; here we extend our computations to derive, for the parameters used, the rainout efficiency values produced by the model. For each case, a semi-empirical relationship between the mass fraction remaining in cloud air, the rainfall intensity and the precipitation time is obtained by using regression analysis of the computational results. The respective equations are:

$$F_1 = 1 - (.044 + .0042\ I)t^{0.90} \tag{19a}$$
(steady state, case I)

$$F_2 = 1 - (.028 + .0017\ I^{1.35})t^{0.68} \tag{19b}$$
(non-steady state, case II)

$$F_3 = 1 - [(.044 + .0042\ I)t^{0.90} + .08] \tag{19c}$$
(steady state, case III)

$$F_4 = 1 - [(.028 + .0017\ I^{1.35})t^{0.68} + .08] \tag{19d}$$
(non-steady state, case IV)

where F is the mass fraction remaining in cloud air; I rainfall intensity (mm/hr); t precipitation time (hr). With these equations, the rainout efficiency for each case can be expressed as:

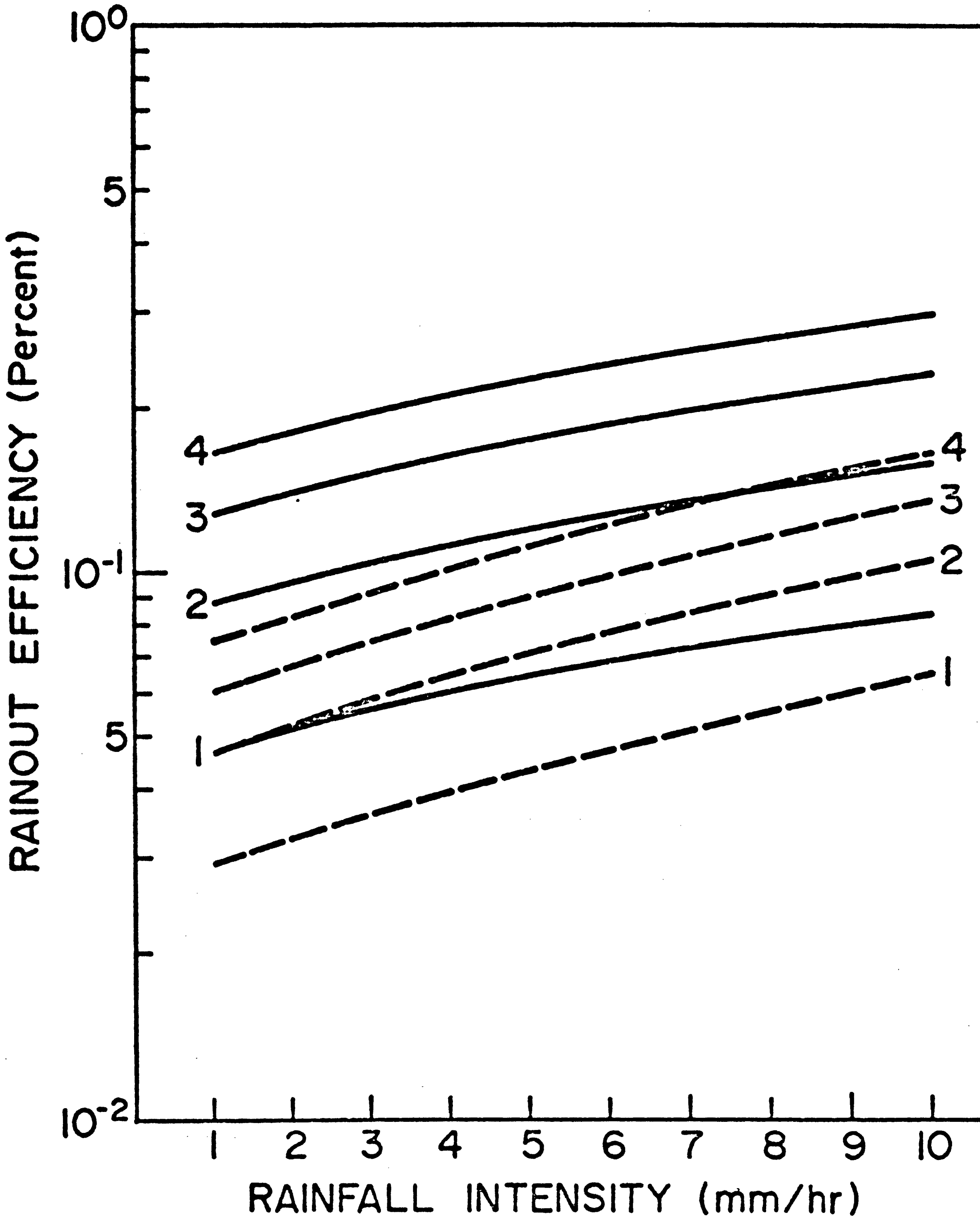

Fig. 9. Rainout efficiency vs rainfall intensity for Case I (solid) and Case II (dashed). Precipitation time in hours is indicated by the numbers.

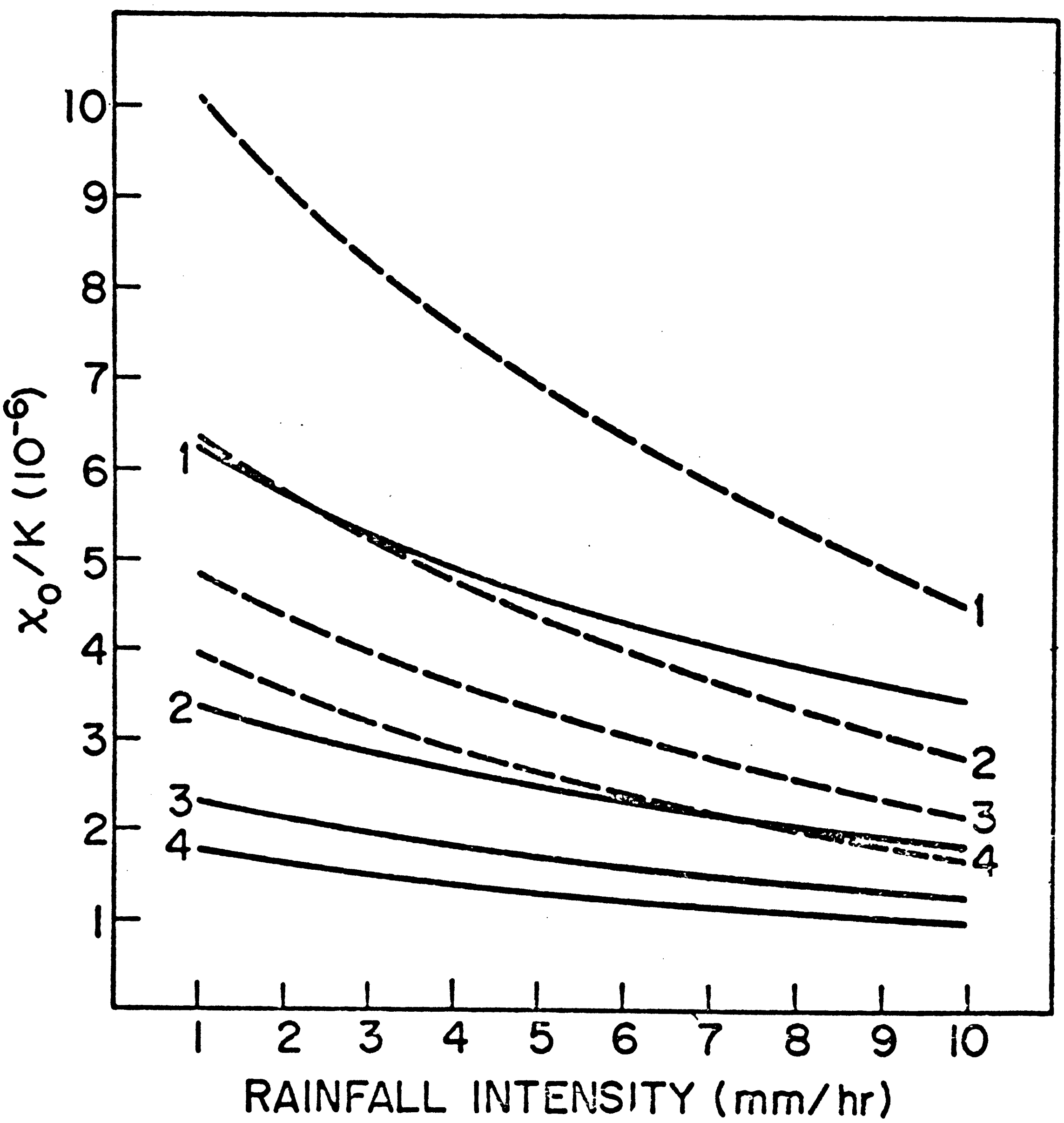

Fig. 10. χ_o/K vs rainfall intensity for Case I (solid) and Case II (dashed). Precipitation time in hours is indicated by the numbers.

Table 2: SELECTED MEASUREMENTS OF SURFACE AIR TO PRECIPITATION ACTIVITY RATIOS AND VALUES OF CLOUD-WATER CONCENTRATION CHOSEN TO PRODUCE RAINOUT EFFICIENCIES BETWEEN 0.5 AND 1.0

(after Engelmann, 1968)

DATA SOURCE	χ_0/K	K/χ_0	Q/ρ_ℓ	E
Small* (Norway)				
October 1956	0.25×10^{-6}	4.0×10^{6}	0.25×10^{-6}	1.0
September 1959	2.06	0.47	2.0	0.95
Average (3 year)	0.9	1.1	0.9	1.0
Hinzpeter (Germany)				
Rain, mm/day				
0.1	0.8	1.25	0.5	0.62
1.0	1.4	0.71	1.0	0.71
10.0	2.5	0.40	2.0	0.8
Snow, mm/day (water equivalent)				
0.15	0.9	1.1	0.5	0.55
1.0	1.6	0.62	1.0	0.62
10.0	3.4	0.29	2.0	0.59

*
S. H. Small, 1960

Hinzpeter, 1958

$$E_1 = 1 - F_1 = (.044 + .0042\ I)t^{0.90} \quad (20a)$$

$$E_2 = 1 - F_2 = (.028 + .0017\ I^{1.35})t^{0.68} \quad (20b)$$

$$E_3 = 1 - F_3 = (.044 + .0042\ I)t^{0.90} + .08 \quad (20c)$$

$$E_4 = 1 - F_4 = (.028 + .0017\ I^{1.35})t^{0.68} + .08 \quad (20d)$$

Engelmann (1971) defines a "washout ratio" in terms of K/χ_0 (in the present context, the term "rainout ratio" appears more appropriate) and strongly recommends utilizing the ratio for predicting in-cloud scavenging of bomb debris.

Introducing E_i into Eq. (18), we have

$$\frac{K}{\chi_0} = E_i \frac{\rho_\ell}{Q} \quad (21)$$

This shows that the rainout ratio varies directly with precipitation rate and duration of rainfall. With reasonable value of Q, here $Q = 3.0 \times 10^{-7}$ gm/cm^3, the rainout efficiency and the rainout ratios can be determined. Fig. 9 and Fig. 10 show these results which are compatible with experimental observations (see Table 2).

Referring to Fig. 9 we can see that for various rainfall intensities and precipitation times, the rainout efficiency is in the range of 0.03 - 0.3. In view of the semi-empirical expressions deduced for different cases, we may suggest a more general expression for the in-cloud rainout efficiency as

$$E = (E_d + a_0\ I^{a_1})\ t^{a_2} + E_n \quad (22)$$

where E_n is the fraction of pollutant serving as condensation nuclei, a_0, a_1, a_2 are constants, E_d can be considered as the fraction of pollutant attached to precipitation elements due to diffusive processes, and the meteorological parameters I and t can be well determined. With reasonable values of these parameters, Eq. (22) may be used as quantitative indications of in-cloud scavenging wherever observations or data source are inadequate.

2.4 SUMMARY

The parameters introduced into this model are based on the elementary consideration of physical processes and take into account particle and droplet size spectra. In previous studies, Greenfield (1957) proposed the theory of scavenging of contaminant on the basis of monodispersed particles and droplets. Makhon'ko (1967) and Davis (1972) have deduced the order of magnitude of these parameters (α, λ) from the ground experimental data. Within the limits of our model, the order of magnitude, shown by these computations, is 10^{-5} to 10^{-4} for α, 10^{-6} to 10^{-5} for β, and 10^{-4} to 10^{-3} sec^{-1} for λ, which compare favorably with the experimental results. The value of γ is found to be negligible as noted above. The case of constant cloud water (Cases I & III) may be considered as an upper limit for the fraction scavenged, while on the other hand, the case of reducing cloud water (Cases II & IV) may be considered as a lower limit.

The removal rate pattern is closely related to the time of residence of the contaminant in the cloud region before precipitation starts and also to the rainfall rate. The method and the results provide quantitative evidence of the relative importance of the different scavenging mechanisms.

The parameters involved in the general expression Eq. (22) can be varied over some limits in order to determine the effects of processes being parameterized. The estimates of E show that diffusive processes are important factors of in-cloud scavenging. In the model we propose to represent the in-cloud scavenging processes. It is recognized that electrical charge distributions in clouds are not well understood, and that the rates of energy dissipation by relevant frequency bands in the turbulence spectrum are also inadequately known, while the phoretic processes contributing to in-cloud scavenging remain somewhat uncertain. We propose here a model that appears to be of general validity and will continue to hold when these physical inputs to the system are more adequately treated.

CHAPTER 3

A MODEL FOR VARIATION OF CONTAMINANT CONCENTRATION WITH RAINDROP SIZE IN CONVECTIVE STORMS

It has long been observed that in convective rains, the variations of contaminant concentrations in successive rain samples are opposed to synchronous variations of rainfall intensity, but the heaviest concentration in the heaviest rain also occurs at times. Mechanisms such as nucleation, coalescence and evaporation have been suggested to explain inverse relationships of rainfall rate to concentration of contaminant. (Huff and Stout, 1964; Bleeker, et al., 1966; Hicks, 1966). A number of paper (Kruger and Hosler, 1963; Huff and Stout, 1964, etc.) have also suggested that the processes which produce direct relationship must be distinct or different from those which produce the inverse relationship. However, there is no general agreement on the cause of either the direct or inverse relationship.

Previous investigations of the "direct" and "inverse" relationships have been concentrated on the interpretation of the causes of the correlation of contaminant concentrations in sequential rain samples to the rainfall intensity. However, the above analyses are not extended to numerical experiments, and the relation of the mass of any specified contaminant to the pure water of an individual raindrop or cloud droplet as the drop grows or evaporates in or below a a convective cloud deserves further study. In the present study, we shall therefore confine our attention, for the most part, to the precipitation elements within a cloud. The discussion is focused on the equation of coalescence growth under certain assumptions about cloud properties and on the equation of the horizontal motion of raindrops. The contaminant concentration in each individual raindrop in the growing or evaporating phase is also modeled. The preliminary results presented here support the descriptive model proposed by Gatz and Dingle (1971) and may shed a little light on concentration variations during convective rains as well as on the physical mechanisms responsible for producing the two types of relationships.

3.1 The Model

3.1.1 Updraft Profile

In order to have a completely compatible model which accounts for the cloud formation and motion, it is necessary to solve a non-linear non-stationary system of equations, including equations of heat and moisture balance as well as equations of motion and continuity. Such a full scale solution is beyond the scope of this study. For reasons of simplicity, some updraft profiles which are considered to be valid and can approximately describe the vertical motion in a cloud system are introduced.

In prior treatments of cloud convection, a "top hat" profile (Squires and Turner; 1962, Srivastava and Atlas; 1969) has been assumed. That is , one in which all properties under consideration are taken as uniform across the updraft and zero outside. In this study, a Gaussian profile (Goldman, 1968) is considered as a more realistic representation of updraft in convective clouds.

The assumed updraft profile is specified by the following analytical expression:

$$W = 0.91\, W_{max} \left(\sin \frac{\pi z}{H} - \frac{1}{4} \sin \frac{2\pi z}{H}\right) e^{\frac{(\frac{-y^2}{2})}{2y_s}} \qquad (23)$$

where W is the vertical velocity, z and y denote the vertical and radial coordinates respectively, H is the height of the cloud top, y_s stands for the distance scale, and W_{max} represents the maximum vertical velocity. The expression in brackets is adopted from Fujita and Grandoso (1968).

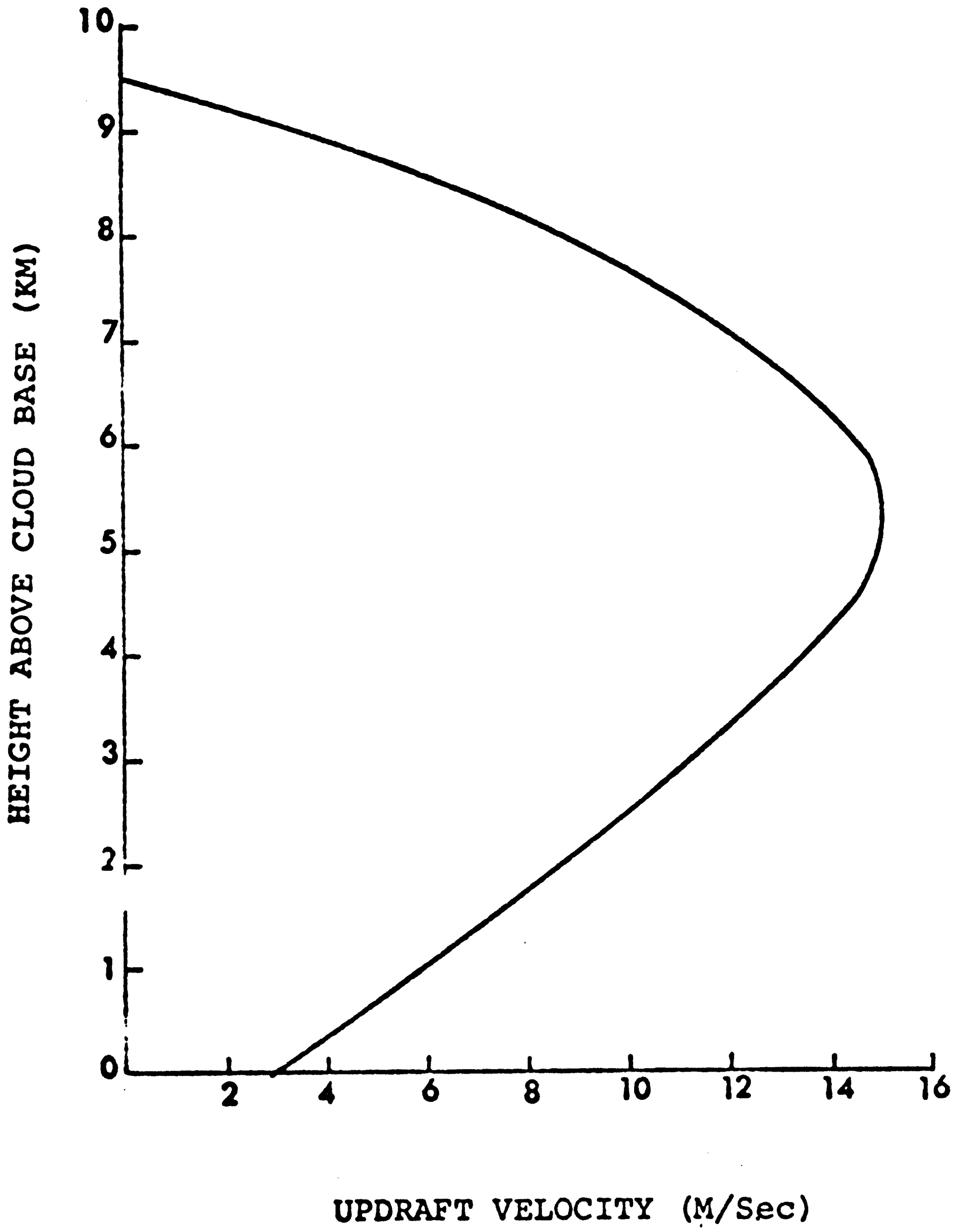

Fig.11. Updraft profile

(after Fujita and Grandoso,1968)

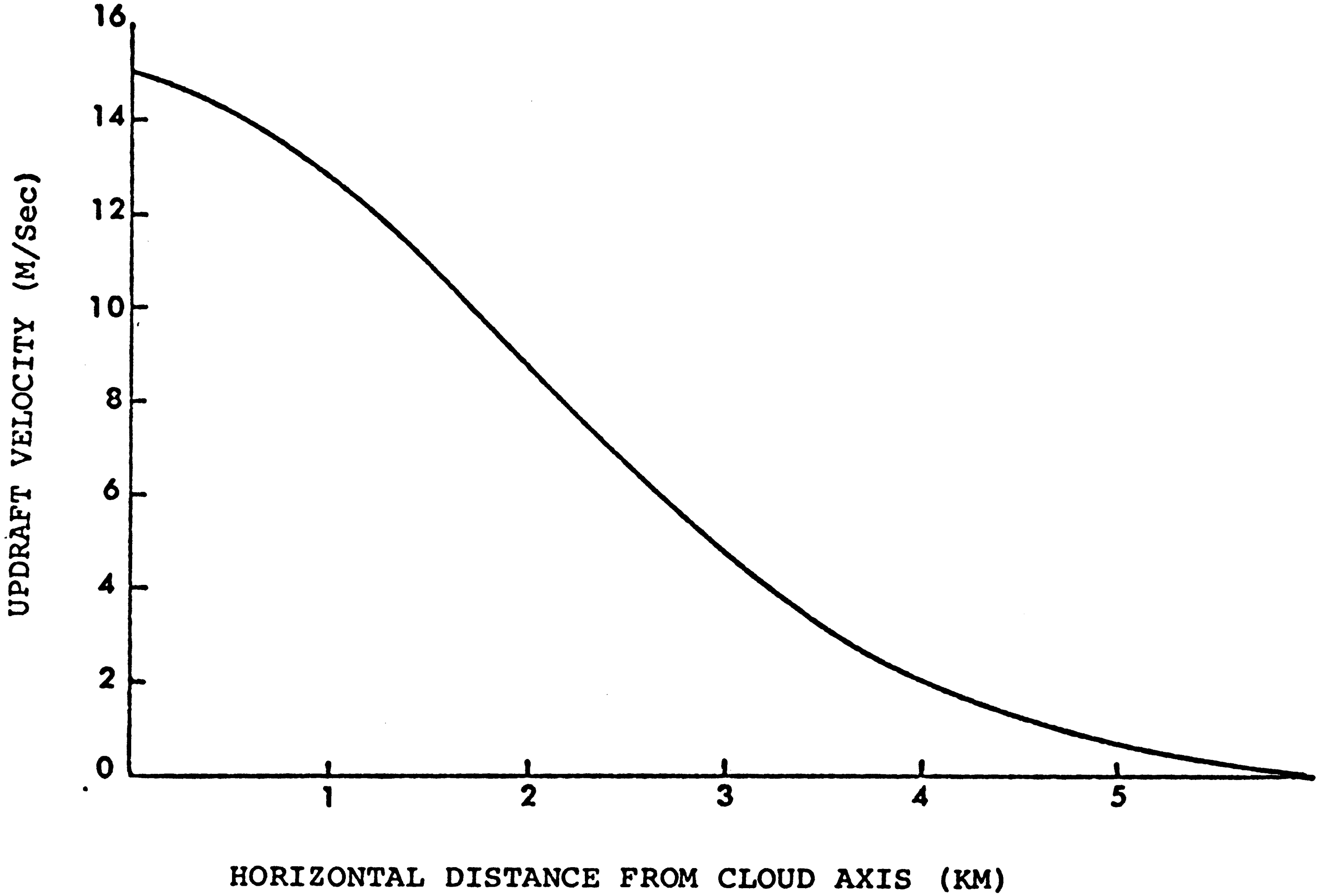

Fig. 12. Horizontal profile of the updraft at 6 km above cloud base.

With this profile, we will have the following characteristics of updraft in cloud: i) the updraft increases with height and peaks at the level $z \approx 0.62H$ (Fig. 11), ii) to consider the eroding effects on the velocity profile by mixing due to entrainment, the horizontal profile of the updraft is assumed to be bell shaped (Fig. 12), iii) the form of the horizontal variation is considered to be axially symmetric so as to maintain mathematic simplicity of the problem.

3.1.2 The Growth Equation

Following Mason (1971), the coalescence growth rate of drops is described by

$$\frac{dR}{dt} = \frac{\pi}{3} \int_o^R n(r)\ r^3\ E(r,R)\ (V_R - V_r)\ dr \qquad (24)$$

where t is the time. To make the integration easier, we assume that E(r,R) is a constant in each time step, substitute (15), in Eq. (24), and after integration, we obtain

$$\frac{dR}{dt} = 2.08 \times 10^{-3}\ E(r,R)\ Q\ \left(V_R I_1 - \frac{2\rho_\ell g \bar{r}^{-2}}{81\eta}\ I_2\right) \qquad (25)$$

where

$$I_1 = 120 - e^{-bR}\ (R^5b^5 + 5R^4b^4 + 20R^3b^3 + 60\ R^2b^2 + 120\ Rb + 120).$$

$$I_2 = 5040 - e^{-bR}\ (R^7b^7 + 7R^6b^6 + 42R^5b^5 + 210R^4b^4 + 840\ R^3b^3 + 2520\ R^2b^2 + 5040\ Rb + 5040).$$

and b is defined in equation (15).

The liquid water content Q is assumed to increase linearly from the cloud base to cloud top (Srivastava and Atlas, 1969) and to decrease with the distance from the cloud axis due to entrainment. Q has the form

$$Q = Q_o\ (z - z_o)\ e^{\left(-\frac{y^2}{2y_s^2}\right)} \qquad (26)$$

where Q_o is a constant and z_o is the cloud base height.

3.1.3 Horizontal Motion

Following Srivastava and Atlas (1969), the horizontal motion of precipitation elements inside the cloud can be described by the continuity equation. The requirement of continuity in an axially symmetric system is

$$\frac{1}{y}\frac{\partial}{\partial y}\ (\rho_a y u) + \frac{\partial}{\partial z}\ (\rho_a w) = 0 \qquad (27)$$

where u is radial velocity of the air and ρ_a is the air density. Concerning the vertical distribution of air density, the hydrostatic equation and the ideal gas law lead to

$$\rho_a = \rho_o\ [1 + \frac{\Gamma z}{To}]^{-(g + \Gamma R_a)/R_a \Gamma}$$

where ρ_o is the air density at ground level; Γ is the vertical gradient of temperature T and is assumed to be nearly constant; T_0 is the surface temperature; g is the acceleration due to gravity; and R_a is the gas constant for air. For values of Γ representative of the real atmosphere, $\frac{\Gamma z}{To} \ll 1$, and therefore, $1 + \frac{\Gamma z}{To} \approx e^{\frac{\Gamma z}{To}}$. Then the above equation can be written as:

$$\rho_a = \rho_o\, e^{-\left(\frac{g + \Gamma R_a}{RT_0}\right)} = e^{-\delta z} \quad \text{(Kessler, 1969)} \qquad (28)$$

where $\delta = \frac{g + \Gamma R_a}{R_a T_o}$. In the standard atmosphere below the isothermal layer, $T_o = 288^0$ K, $\Gamma = 6.5°c/km$, $\delta = 0.96 \times 10^{-6}$ cm, but the line connecting densities at z=0 and z=10^6 cm in semi-log coordinates has a slope corresponding to $\delta = 1.09 \times 10^{-6}$ cm^{-1}. In the calculations presented in this study, $\delta = 10^6$ cm^{-1} (Kessler, 1969). Using (23) and (28), Eq. (27) gives

$$\frac{1}{y}\frac{\partial}{\partial y}(yu) = \left[-0.91\, W_{max} \left\{ \frac{\pi}{H}\left(\cos\frac{\pi z}{H} - 0.5\cos\frac{2\pi z}{H}\right) - \delta\left(\sin\frac{\pi z}{H} - 0.25\sin\frac{2\pi z}{H}\right)\right\}\right] e^{\frac{-y^2}{2y_s^2}}$$

$$= f(z)\, e^{\frac{-y^2}{2y_s^2}} \qquad (29)$$

where f(z) represents all terms in the brackets.

Integrating (29) from the center y = 0, u = 0 to a distance y, yields

$$u = \frac{dy}{dt} = \frac{\sigma^2}{y} f(z) \left(1 - e^{\frac{-y^2}{2y_s^2}}\right) \qquad (30)$$

In the cloud, the true upward velocity of a large drop is

$$\frac{dz}{dt} = W - V_R \qquad (31)$$

3.1.4 Evaporation of raindrops

In treating evaporation of raindrops below the cloud, Liu and Orville (1969) consider the Reynolds number constant. We introduce here consideration of Reynolds number as a function of drop sizes, thus making this term more complete than in the prior treatments.

The evaporation for a single drop, assuming thermodynamic equilibrium, is

$$\frac{dR}{dt} = \frac{(S-1)\ (1+F_v Re^{1/2})}{R(A_o+B_o)} \qquad \text{(Byers, 1965)} \qquad (32)$$

with

$$A_o = \frac{L^2 \rho_\ell}{K_t R_v T^2}$$

$$B_o = \frac{T\ R_v\ \rho_\ell}{P_o\ (T)\ D_f}$$

where S is the ambient saturation ratio, F_v the ventilation factor and has the value 2 for Re $\geq$ 5.6 (Abraham, 1968), L the latent heat of vaporization, K_t the thermal conductivity of air, R_v the gas constant for water vapor, T the ambient temperature, P_o (T) the saturation vapor pressure at T, and D_f is the diffusivity of water vapor. The Reynolds number, Re, can be related to the raindrop sizes using regression analysis of the data observed by Gunn and Kinzer (1949).

The relationship is

$$Re^{1/2} = - 0.4930 + 371.0999\ R - 775.2944\ R^2 + 72.2532\ R^3 \qquad (33)$$

In the case of evaporation, the numerator of Eq. (32) would be negative because the saturation ratio S is less than 1. This ratio in percentage form is the relative humidity and an important variable in determining dR/dt (in our computations the relative humidities are varied, see Fig. 16). In the denominator, the two terms A_o and B_o are of the same order of magnitude (10^5 sec cm^{-2}) over a temperature range between 10° c and 30°c; A_o is greater than B_o, but B_o increases more rapidly than A_o as temperature decreases.

3.1.5 Terminal speed correction

The equations of terminal fallspeed, (Dingle and Lee, 1972), are applicable near ground level. To apply them to other levels, a correction factor should be introduced for the effect of air density on terminal fallspeed. The expression

$$V(Z) = V(O)\ \left(\frac{\rho_o}{\rho_a}\right)^{0.4} \qquad \text{(Foote and Du Toit, 1969)} \qquad (34)$$

previously used by Atlas, et al (1971) in the study of the doppler radar reflectivity spectrum is used in this study; V(O) is the droplet fallspeed at ground level and V(Z) is the fallspeed at the level of observation.

3.1.6 Raindrop Breakup and Chain Reaction

Experiments carried out by Blanchard (1948, 1950, 1962), by Koenig (1965), by Cotton and Gokhale (1967), and by Beard and Pruppacher (1969) have shown that drops of radius larger than a certain size become unstable and break into smaller drops. In the present study, we assume that 0.275 cm is the maximum allowable radius for raindrops. Drops reaching this critical size are broken into a few larger drops of millimeter size, together with a large number of very small droplets. In the computation of drop growth, it is assumed that if breakup occurs, at least one of these fragments has a radius 0.173 cm. A drop of this radius has about one-fourth mass of the drop of original radius 0.275 cm. These fragments are rising and falling or balanced ($V_R = W$) within the updraft depending on their horizontal and vertical positions relative to the cloud axis and their fall velocities.

3.1.7 Contaminant Distributions

Evidence (Dingle and Gatz, 1966; Huff, 1965; Small, 1960) shows that contaminants in most clouds can be considered as originating from low-level sources. Considering then the low-altitude input in conjunction with such sound physical processes as the selective activation of condensation nuclei, capture of particles by cloud and/or raindrops in rising air, and the effects of both turbulent diffusion and the updraft profile, it is evident that those mechanisms must generate both a vertical and horizontal gradient of cloud droplet solution concentrations. The vertical gradient of the contaminant concentration extends from a maximum near the cloud base to a minimum in the upper part of the cloud, while the horizontal gradient of the contaminant concentration, has its maximum at the core of the updraft.

Because of the complexity of convective storms, the complete description of contaminant distribution and theoretical evaluation has long been an elusive objective of precipitation scavenging research. We assume that the contaminant concentration in cloud water may be expressed by:

$$C(Y,Z) = C\ (Y_o,Z_o)\ e^{-\left(\frac{Y^2}{2y_s^2} + \frac{Z^2}{2z_s^2}\right)} \tag{35}$$

where $C(Y_o,Z_o)$ is the contaminant concentration (mass of contaminant/gH_2O) at the cloud base and, y_s and z_s are distance scales for Y and Z coordinates respectively. As the raindrop grows by coalescence with cloud droplets, the mass of contaminant acquired by a raindrop can be written as

$$M_c(Y,Z,t) = \frac{4}{3}\pi\rho_\ell \{ R^3\ (Y_2,\ Z_2,\ t_2) - R^3\ (Y_1,\ Z_1,\ t_1)\}\ c\ (Y,Z) \tag{36}$$

where the subscripts refer to two different time steps. Eq. (36) estimates the amount of contaminant mass scavenged by a raindrop during its growth in a storm.

3.2 Model Calculations

a) Numerical methods. The set of equations (25), (30), (31) and (32) was numerically integrated. This involves the replacement of the time derivatives of R, Z, and Y with a set of finite differences. In doing so we obtain approximate values for the foregoing dependent variables at a set of discrete points. There are various finite difference schemes that could be used. In this study we have chosen to use a RUNGE-KUTTA method of fourth order accuracy. In using numerical methods, it is necessary to investigate 1) truncation error, and 2) stability. Here the truncation error is $O(h^5)$, where h is the time step. The time step used in the calculations was 10 seconds. A check on the accuracy of the method was made by halving the time step and repeating the calculations. No significant differences were found between the two solutions. For a complicated set of equations, it is not possible to derive a general stability criterion. In this case the stability can only be tested empirically. Our computations show that the results from 5 second time steps agree favorably with the results from 10 second time steps at each time step. Thus we may say that the numerical method will be stable.

b) Parameters. The important parameters for the quantitative description of physical inputs to the model cloud are i) cloud water content, ii) maximum vertical velocity, and iii) initial horizontal entry positions. In our numerical analysis of the model, these three parameters were varied in order to determine its effect on the precipitation growth and motion.

c) Cases. Computations were run for

i) three intensities of updraft (W_{max}): 10, 15 and 20 m/sec.

ii) two initial horizontal entry positions: 1 m and 2.0 km from cloud axis.

iii) two Q_o values: 1.5×10^{-6} and 3.0×10^{-6} gm/cm^3.

In computing all these cases, eleven arbitrary sizes of initial radii 30, 40, 45, 50, 55, 60, 65, 70, 75, 90 and 100 μm are introduced into the base of a developing cloud with a base radius of 5 km. A few results of these computations are presented in the next section.

3.3 Results and Discussions

3.3.1 Case I. In this case, we assume $Q_o = 3.0 \times 10^{-6}$ gm/cm^3, W_{max} = 15 m/sec, and all drops enter the cloud very close to the cloud axis (y = 100 cm). It is seen from Eq. (30) that the raindrops on the cloud axis (y=0) do not undergo any horizontal displacement, and the horizontal displacement of a raindrop depends on 1) the maximum vertical velocity (W_{max}), and 2) the shape of the updraft profile.

Examination of the computations show that a drop of initial radius 60 μm, reaches a height about 7.4 km (above the peak updraft) and grows to the critical radius (0.275 cm) in about 24 minutes. Then breakup takes place; one of the larger fragments (R = 0.173 cm) in turn grows in its falling path and breaks up again. Computations show that the breakup process is repeated 18 times in a layer of about 1.7 km depth between 7.4 and 5.7 km. Finally, a drop of radius 0.226 cm, after spending about 96 minutes in the growing and breakup processes, emerges at cloud base at a distance of 1.33 km from the cloud axis. Similarly, a drop of initial radius 100 μm, growing to critical size and breaking up 21 times in about 100 minutes, will eventually fall out of the cloud base with a radius of 0.253 cm after undergoing a horizontal displacement of 1.35 km. Drops of other initial radii, entering the cloud at the same position, show similar behavior. This process popularly known as Langmuir chain reaction has been considered as an important contribution to precipitation formation in warm clouds.

3.3.2 Case II. In this case, we take $W_{max} = 15 \pm$ 5m/sec, the other conditions being similar to Case I. It is seen that the increase in the updraft by 5m/sec, as compared to Case I, causes a drop to breakup sooner and at a higher altitude. Conversely, a decrease in the updraft by 5m/sec tends to cause the fracture to occur later and at a lower altitude under these conditions than under those of Case I.

3.3.3 Case III. In this case, the initial conditions are the same as for Case I and Case II, except that Q_o was taken as 1.5×10^{-6} gm/cm^3, which provides a contrast with Case I and Case II (twice the liquid water content). Computations were run for W_{max} = 10, 15 and 20 m/sec. Results of these computations show that 1) there is relative little breakage, 2) fracture occurs later, as compared to Case I and Case II for each updraft velocity.

In view of the above results, it may be noted that if the drop growth takes place by the suggested mechanism; coalescence and breakup, then a number of consequences may be stated: 1) the breakup mechanism operates to sort the precipitation particles, the smaller drops ($W>V_R$) will move upward with further growth, while the larger drops ($V_R>W$) will grow as they descend, 2) the stronger the updraft and the higher the liquid water content, the sooner the fracture occurs. Consequently, many more drops are produced in the upper regions of a storm, 3) the resulting drops, after under-

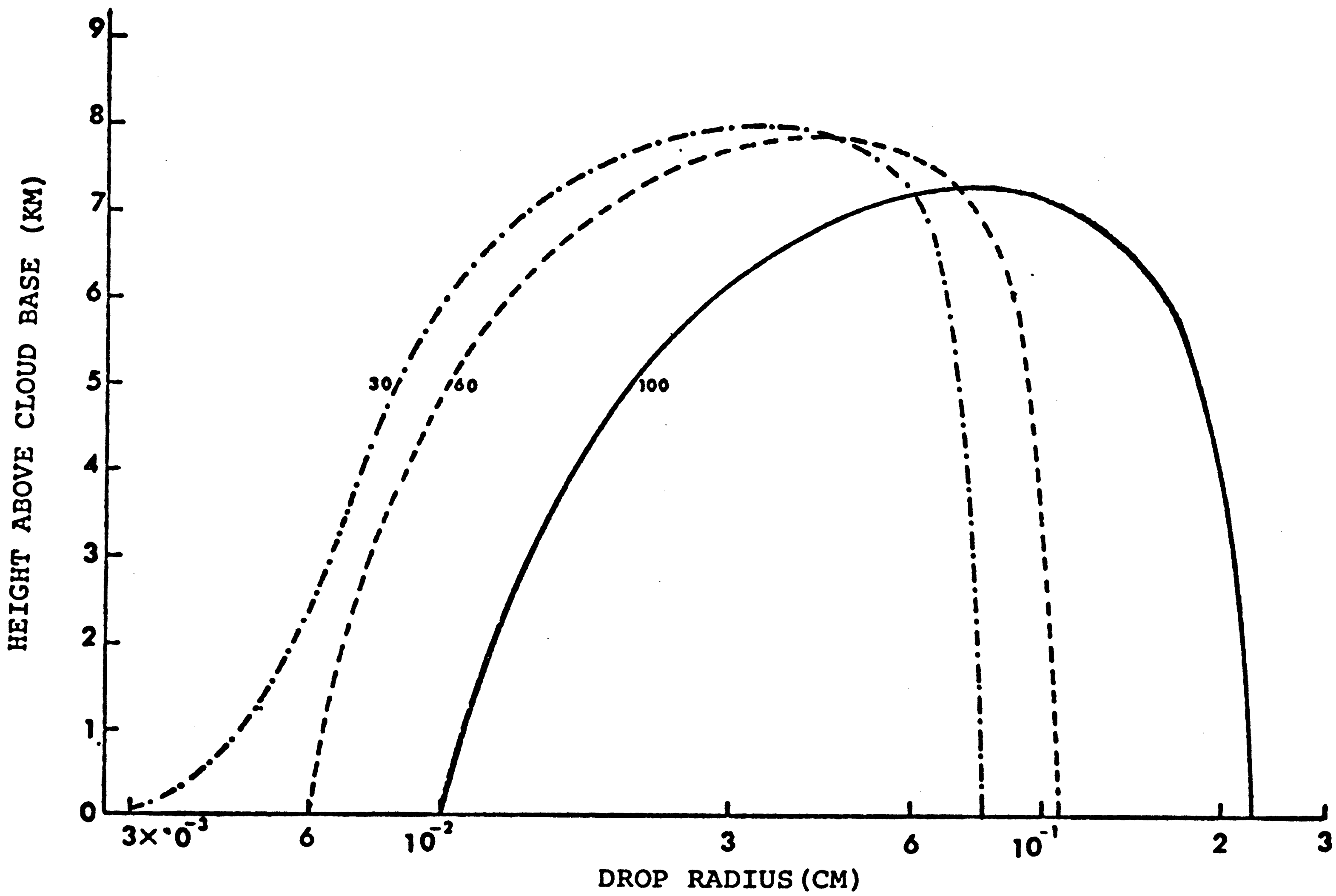

Fig. 13. Change in drop radius with height (Case IV), numbers on curves indicate initial drop radius (micron).

going the same processes as discussed above, grow to millimeter (about 2mm) sizes and emerge from the cloud base in a region near the storm core. These larger drops generate heavy precipitation.

3.3.4 Case IV. In this case, we take $Q_0 = 3.0 \times 10^{-6}$ gm/cm^3, W_{max} = 15m/sec, and all the drops are assumed to start at a distance of y=2 km from the cloud axis. Trajectories of drops which grow by coalescence in a cloud with the assumed "bell-shape" profile were computed. Fig. 13 relates drop radius to height above cloud base. For the sake of clarity, the growth curves only for drops of initial radii 30, 60 and 100μm are shown. As can be seen, in the course of their development, the mass increment of a growing drop on its ascending and descending branches are significantly different. For example, (see Fig. 13) a drop of initial radius of 30μm grows to 332μm as it ascends and reaches a height of 8.0 km. The mass of this drop is increased by a factor of about 1,350. Meanwhile, the same drop turns downward with further growth and emerges from the cloud base with a radius of 784μm, and the mass is increased only by a factor of about 13. Drops of other radii indicate the same features. Evidently, for this case, (Case IV) growth mainly proceeds in the upward passage rather than in the downard passage.

Bowen (1950) performed somewhat similar calculations assuming the liquid water content and the updraft to be uniform throughout the cloud. His results show that the change

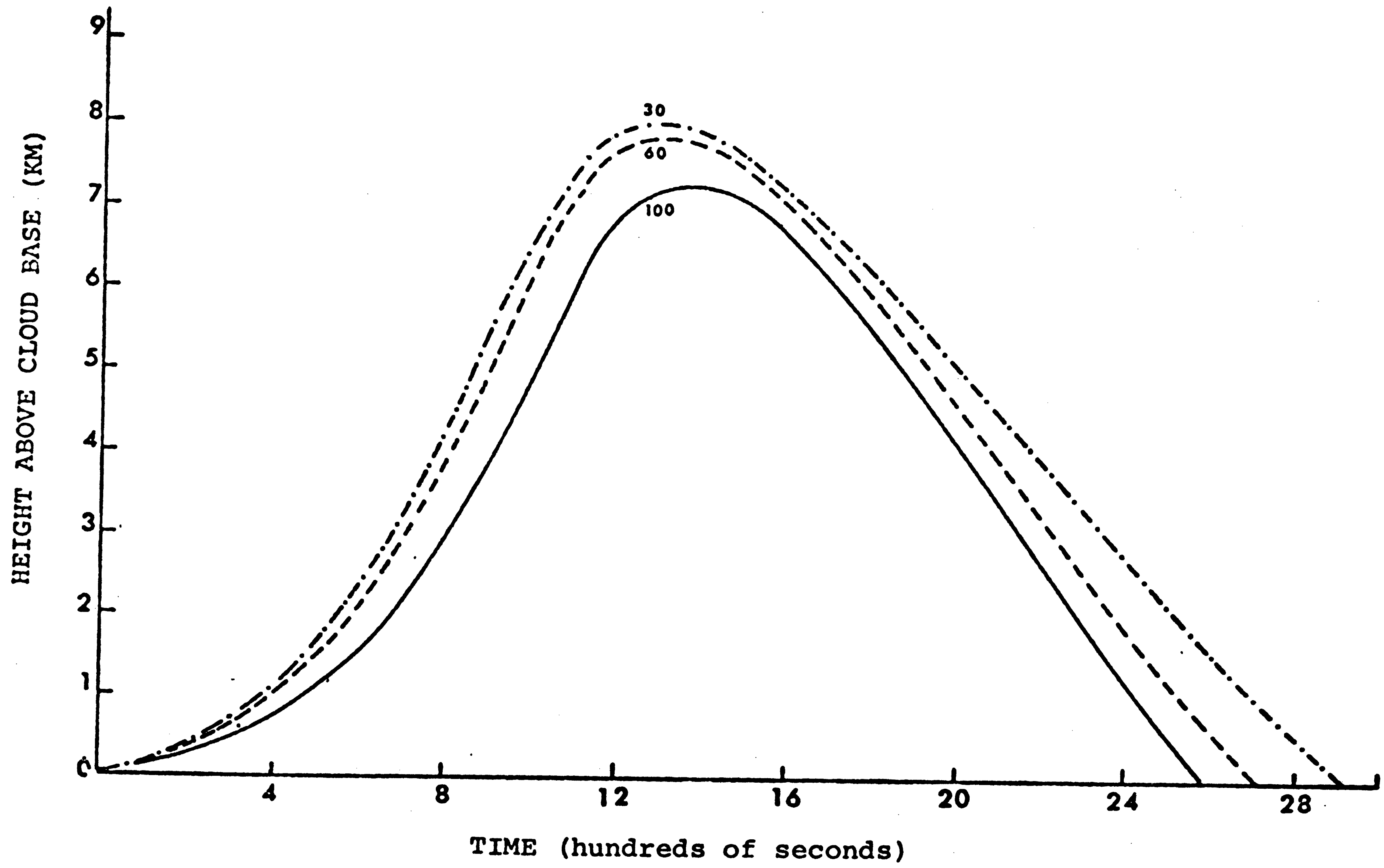

Fig. 14. Time vs height above cloud base for various drop sizes (Case IV), numbers on curves indicate initial drop radius (micron).

in drop radius with height essentially takes place in the downward trajectory rather than in its upward trajectory. Recently, Srivastava and Atlas (1969) have made similar calculations for drops of several arbitrary sizes started at a height of 3 km above cloud base under the assumptions of "top-hat" profile and constant liquid water content through a section at any level. Their results indicate that drops having an initial fall speed less than a "critical" fall speed which they define do not grow very rapidly with height as they move upward. The difference found here, as compared to the previous results (Bowen, 1950; Srivastava and Atlas, 1969), are attributed to the assumed profiles of updraft and cloud water content in the model cloud.

Fig. 14 shows the altitude of growing drops as a function of time. As can be seen from the figure, all the drops spend approximately the same period of time in the growth stages: that is, the initial stage during which the drops are carried by the updraft, and the final stage during which the drops become large enough to fall against the updraft. In this Case a drop of initial radius 100μm spends about 42 minutes in the cloud (see Fig. 14) and falls out of the cloud base with a radius of 0.226 cm (see Fig. 13). While in Case I a drop initially of 100μm radius grows to the critical size (0.275 cm) in about 23 minutes, then the breakup process takes place and yields fragments for repeated growth. If one of these fragments has a radius of

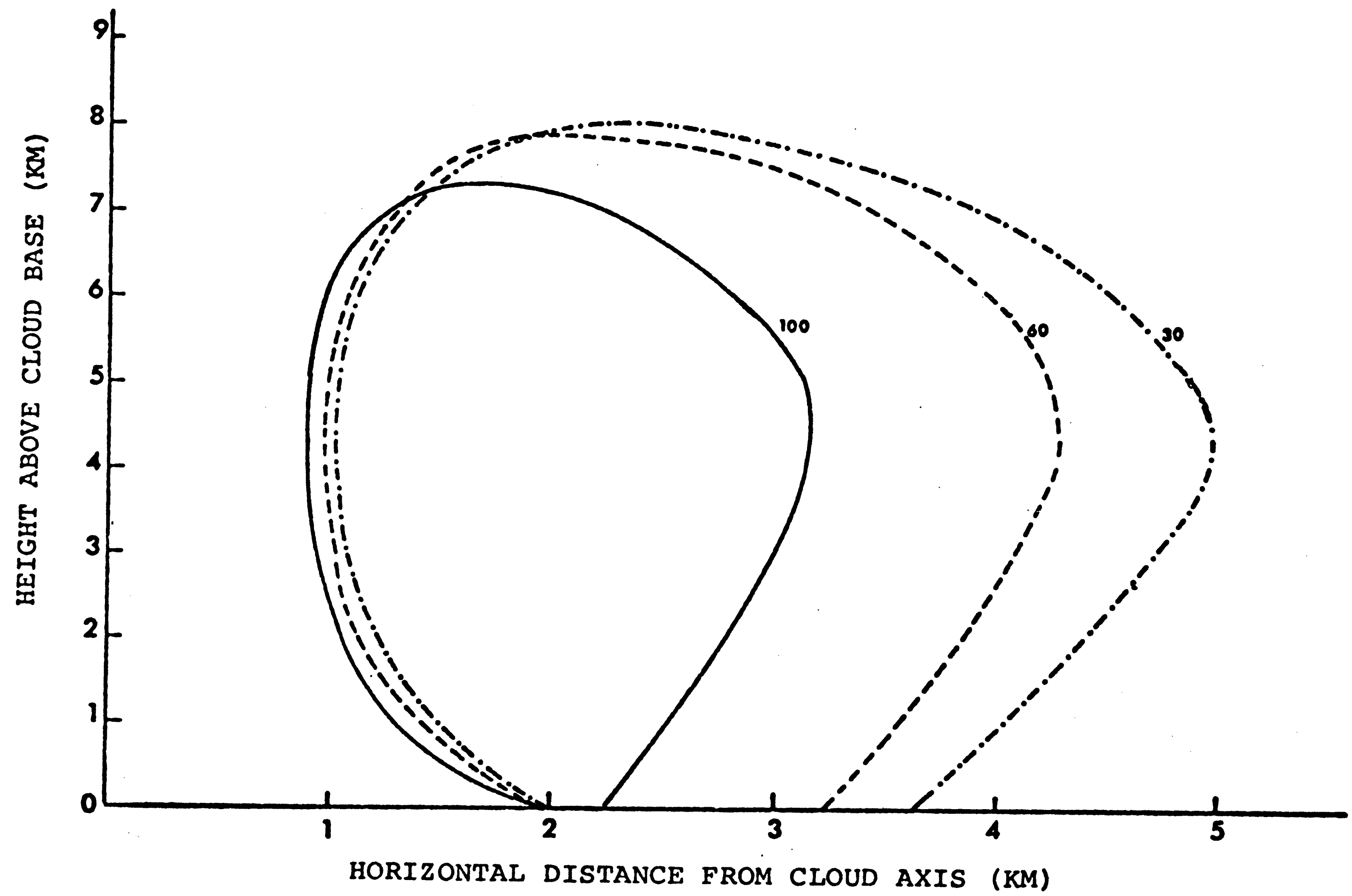

Fig. 15. Trajectories of various sized drops (Case IV), numbers on curves indicate initial drop radius (micron).

0.173 cm, this drop will repeat the cycles until it finally emerges from the cloud base with a radius of 0.253 cm in another 77 minutes.

Contrary to the results found in Case IV, the growth trajectory shown by Bowen (1950) indicated that under the specified conditions the drop growth by coalescence is slow at first. However, as the drop turns downward, growth proceeds more rapidly and only about a quarter of the total growth time is taken up as it descends to the cloud base. Srivastava and Atlas' calculations (1969) showed that the time taken by a drop in both its rising and falling path depends on the drop size involved. The smaller drops spend a shorter period of time in the upward trajectory than in the downward trajectory.

The horizontal position of the raindrops against their vertical position is shown in Fig. 15. This figure illustrates the 2-dimensional trajectory of drops within a convective cloud under the combined influence of vertical and horizontal motions. The assumed vertical velocity profile given in Fig. 11 is a typical one for a convective cloud. It implies a pattern of horizontal divergence and horizontal convergence. In the lower part, $\frac{\partial w}{\partial z} > 0$, and in the upper part, $\frac{\partial w}{\partial z} < 0$. The drops are initially carried upwards ($W > V_R$) and inward toward the cloud axis in the region of increasing updraft, then carried away from the cloud axis in the region of decreasing updraft, where the drops rising or falling depends upon their sizes. Finally, they fall through the lower part of the cloud and again are displaced inward. In the divergence zone, the smallest drops are displaced to larger distance where they grow in a region of low cloud water content, whereas the largest drops fall closest to the cloud axis where they grow in a region of high water content. In the present case (Case IV), for example, a drop initially of 100μm radius, after completing the growth trajectory in the cloud without breakup, emerges at cloud base at a distance of 2.32 km from the cloud axis and the final radius of the drop is 0.226 cm. On the other hand, when a drop of initially 30μm radius falls out from a cloud base at a distance of 3.66 km, its final radius is 0.78 cm--a raindrop of medium size. Drops having an initial radius less than 30μm may fall outside the cloud into environmental dry air, where the growth ceases and evaporation takes place; thus they may be completely or partially evaporated during their fall through a great depth of dry air.

At this point it is interesting to note the significant difference between Case I and Case IV. In Case I, all the drops initially introduced into the cloud near the core ($y \approx 0$) break up. Thus many small drops are produced through breakage and taken up in the updraft. Consequently, the supply of drops to the upper region of the storm cell is increased. Evidently, an intense updraft and high liquid water content tend to speed up the production of a large drop population.

On the other hand, as shown in Case IV, none of the drops break up, all of the drops complete their growth path in the cloud and fall directly out of the cloud base.

3.3.5 Case V. Similar to Case IV except that the maximum updraft velocity (W_{max}) was increased and decreased by 5m/sec. As a result of the increase (W_{max} = 20m/sec) in the updraft, drops are carried away horizontally to larger distances from the cloud axis. For example, in this case, a drop of initial radius 100μ, growing to a radius of 0.108 cm, emerges from the cloud base at a distance of 3.58 km from the cloud axis. In Case IV, a drop of the same initial radius grows to 0.22 cm and comes out of the cloud base at a distance of 2.22 km. This is not unexpected, since a stronger updraft accompanies stronger divergence which in turn carries the drops farther away horizontally from the cloud core. The 5m/sec decrease in the updraft has the same effect as bringing the small drops outside the storm. The decrease also results in the breaking up of the larger drops. An interesting point brought out by these computations is that under certain conditions, such as $Q_o = 3.0 \times 10^{-6}$ gm/cm^3, and W_{max} = 15 m/sec, drops could maintain their integrity without breakage and complete their growth path in the cloud. Under different conditions of liquid water content and updraft, breakup occurs and small drops can be displaced to larger horizontal distances in the anvil of the cloud.

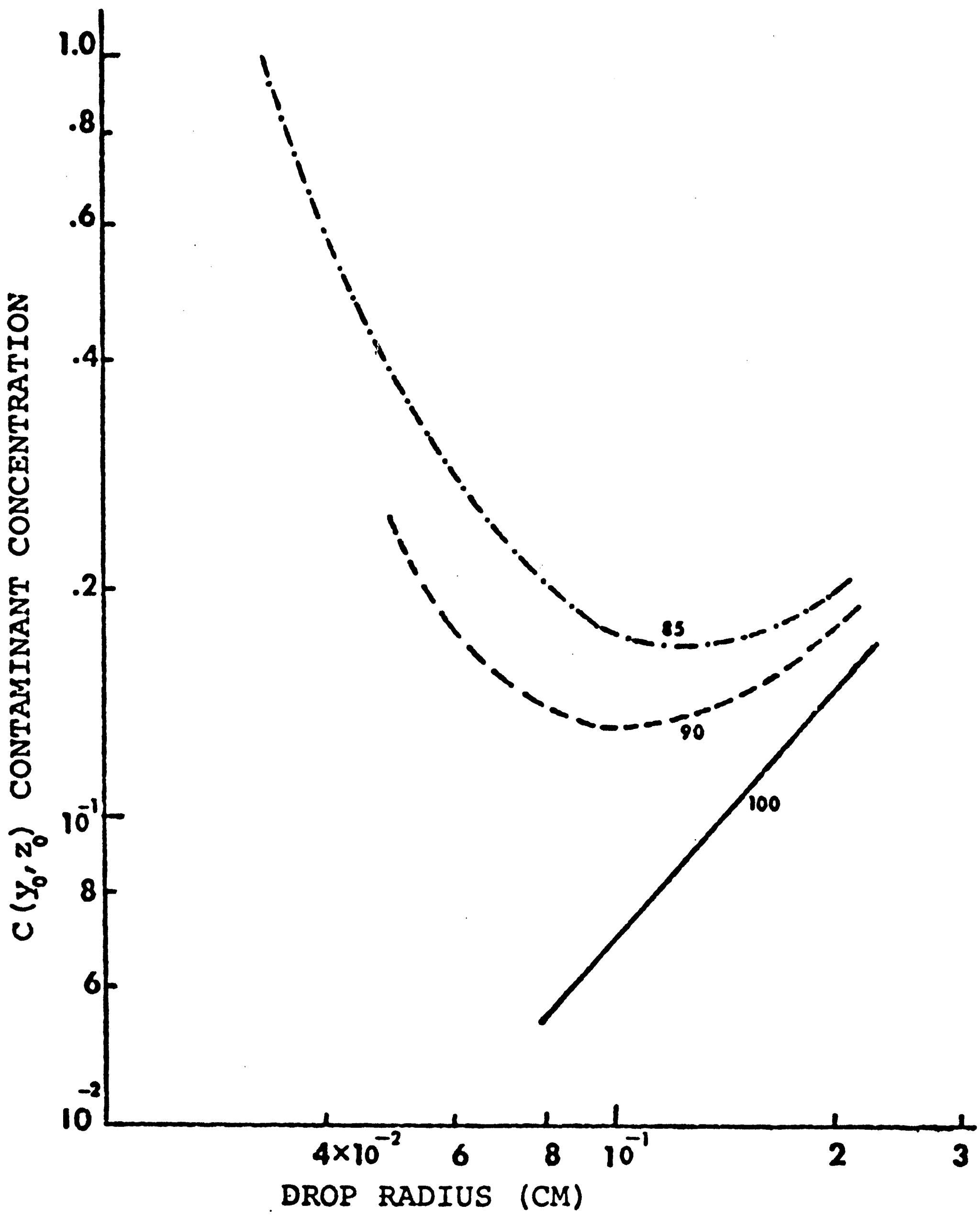

Fig. 16. Relationship between the contaminant concentration of raindrops and their size at various relative humidities (Case IV), numbers on curves indicate relative humidity. $C(y_0, z_0)$ is the initial contaminant concentration (mass of contaminant/g H_2O) at the cloud base.

3.3.6 Case VI. Similar to Case IV, except that $Q_o = 1.5 \times 10^{-6} gm/cm^3$. Calculations were made for $W_{max} = 10$, 15, and 20 m/sec. Results of these calculations are qualitatively similar to those of Case IV and Case V.

3.3.7 Contaminant Concentration - Raindrop Size Relationships

In this section, we shall mainly discuss phenomena which are important in analysing the changes of concentration in the individual drop. By this means we may further the understanding of the variations in the correlation between contaminant concentrations in rain samples and rainfall rates. The precipitation elements which have attained a certain minimum size grow predominantly by coalescence with cloud droplets. In the course of analysis, we consider only the processes of coalescence and evaporation which mainly contribute to variations of contaminant concentrations in the individual drop. Other mechanisms are not accounted for.

The contaminant mass acquired by a growing drop in each time step can be computed by using Eq. 36, whereby the contaminant concentration of individual drops can be determined. Fig. 16 shows the contaminant concentration as functions of drop sizes at the cloud base for Case IV. As discussed in the previous section, larger drops travel lower and closer to the storm core, while smaller drops rise higher and travel farther away horizontally. Thus under the specified condition of Gaussian distribution of contaminant concentration, larger drops become more contaminated, while smaller drops become less contaminated. Therefore the contaminant concentrations are positively correlated with drop sizes. When the drops emerge from the cloud base and fall into the unsaturated environment, evaporation comes into effect and plays an important role in the change of the profile shape. In Fig. 16, some significant features appear. The first of these is that drops of different sizes could have the same contaminant concentrations. The second feature is that it appears there is a critical size for which the drop has a minimum contaminant concentration. Concentrations and drop sizes are positively or negatively correlated depending upon whether or not their sizes are smaller or larger than the critical size. Thus the effect of evaporation is responsible for varying the correlation more or less continuously.

We have shown profile shapes of contaminant concentrations in individual raindrops in relation to their sizes at various relative humidities (Fig. 16). We may then proceed further to explore whether these results are capable of explaining the cause of the variations of "direct" or "inverse" relationships (relation of concentration in a rain sample to rainfall rate).

As the results show, the basic form of precipitation size distributions is established in the cloud, raindrops ranging from R = .078 cm to R = 0.226 cm (Fig. 13) are falling from a ring of radial distance of about 1.4 km (Fig. 14).

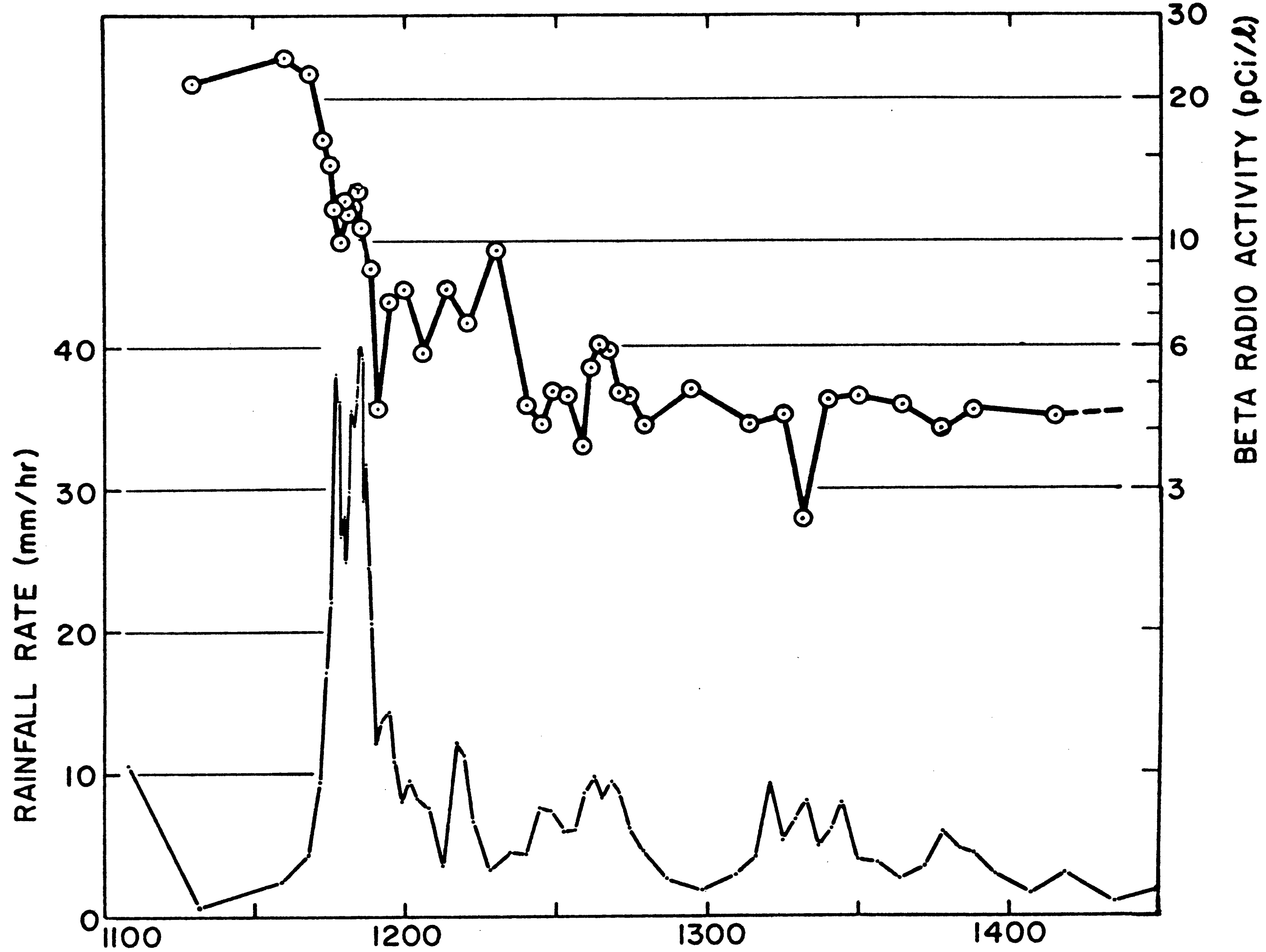

Fig. 17. Rainfall intensity and β-radioactivity concentration profiles for rain of 22 April 1966 at Chickasha, Oklahoma (after Dingle, 1968). Abscissa is CST.

Because of the horizontal sorting of precipitation elements in the cloud region, the largest drop emerges from the cloud base closest to the storm core, whereas the smallest drop is farthest from it.

Thus under the Case IV conditions the result of 1) the size-distribution with different contaminant concentrations spreading in a radial distance of about 1.4 km at the cloud base and 2) the evaporation effect in the sub-cloud region, it is reasonable to predict that a sequence of rain samples collected at ground level may show either an inverse relationship due to the collection of those small drops (a higher concentration in period of low rainfall rate), or a direct relationship owing to the collection of relatively large drops.

Fig. 17 is the observation data of April 22, 1966 (Dingle, 1968). The two principal rate peaks were 5 minutes apart. The radioactivity concentrations were higher at the beginning of the rain and dropped sharply to a minimum in the heaviest rain. In a clearly direct relationship, the second concentration peak and the second rainfall rate peak occurred together. Here is a case to show that very similar maximum rainfall rates occuring in rapid succession at the same place have different relationships. Notice that the shapes of both concentration profiles are typical.

In view of the computation results shown in Fig. 16 and the observation data shown in Fig. 17, it is appropriate to point out that the "direct" or "inverse" relationships are not necessarily produced by different kinds of physical processes.

3.4 Summary

In this chapter, we have obtained several results of basic importance to the growth and motion of precipitation elements in the cloud region, the modification of raindrops due to the effect of evaporation under different humidity conditions, and the variations of concentrations as functions of drop sizes, under certain simplifying assumptions.

Contrary to what has been found by previous workers (Bowen, 1950; Avramenko and Makhonko, 1970), the present analysis indicates that situations exist wherein growth of drops mainly proceeds in the upward trajectory rather than in the downward trajectory. This is mainly a result of the fact that liquid water content and updraft are non-uniformly distributed in the model cloud.

Because of the effects of both horizontal and vertical sorting of drops, many of the drops entering the region of divergence may be expected to fall outside of the cloud into drier environment, where evaporation takes place. Consequently, these drops fall through a great depth of drier air to form the lightest portion of the rain and the highest contaminant concentration. This condition is characteristic of the edges of the shower and is supported by many field observations (Dingle and Gatz, 1966; Gatz, 1966).

The violent updraft and high liquid water content tend to speed up the production of a large drop population through the suggested mechanisms and to increase the supply of drops to the upper region of a storm.

The results shown in Fig. 16 are based on Case IV of this restricted model of precipitation growth and contaminant distribution. It is apparent from these results that the distribution of contaminant in a convective cloud is a very important factor in affecting the amount of contaminant in each individual raindrop. Therefore the contaminant profile should receive more direct attention in future investigation of in- and below-cloud scavenging phenomena.

Finally, these preliminary results appear to us to indicate that the various relationships of rainfall rate to concentration of contaminant do not require different input mechanisms nor different kinds of physical processes, but rather tend to depend upon the structure of the convective storm and its interactions with the environment.

CHAPTER 4

SUMMARY AND CONCLUSIONS

This study proposes two simplified numerical models for precipitation scavenging. Chapter 2 contains the model developed mainly for stratiform clouds. In the course of our analysis, the in-cloud scavenging is viewed as a multi-step process, and the parameters introduced into this model are adequately treated on the basis of microphysical processes. In this way, analytical solutions are obtained which have the advantage of keeping an inventory of the particle sizes as well as giving considerable accuracy in the rainout calculations. On the other hand, the in-cloud scavenging, by virtue of its many interacting and competing processes, is a rather complex phenomenon. In view of such complexity we assume expressions which are rather widely accepted. However many aspects of this analysis require further study. The phoretic effects have been recently investigated by Slinn and Hales (1972), but their results can be viewed as only qualitative indications of the relative importance of the noted mechanisms. The quantitative estimations of these effects contributing to the attachment rate deserve further study.

In order to more comprehensively assess the consequences of the scavenging problem, future plans should include continued research into the areas of: 1) the initial formation of a cloud droplet distribution through condensation on mixed nuclei and 2) the formation of precipitation-size elements through the combined effects of condensation and coalescence.

Incorporation of the microphysical processes (nucleation, Brownian diffusion, impaction, accretion and phoretic effects) into the suggested scavenging model determine: 1) the amount of materials removed by the precipitation generation system, and 2) the fate of materials within the cloud as a function of time and the spatial and temporal distribution of wet deposition on the earth's surface.

Therefore, a more comprehensive model with the inclusion of these processes from this basic model should be the next undertaking. Nevertheless, this basic model is the first attempt to combine the microphysical effects with the size spectra for the study of in-cloud scavenging.

The model presented in Chapter 3 provides a simplified picture of the development of raindrops. It also estimates the vertical and horizontal motion of drops. It can be used to obtain a quantitative estimate of the influences of various parameters upon the processes, and also to explain the cause of the aforementioned direct or inverse relationships. It is important to recall that this model is restricted to the steady-state. Other assumptions also have some effect on the results and it is desirable to refine the model by eliminating some of these restrictions. Thus immediate considerations for future work include the following: 1) treating the updraft and downdraft as functions of both time and space by means of appropriate parameters, 2) evaluating the trajectories of raindrops in the environment, and 3) modelling the contaminant distribution in a convective cloud by considering condensation processes and other microphysical mechanisms mentioned above. Conclusions drawn from results of this study are summarized in the following paragraphs.

The main conclusions which follow from the work described in Chapter 2 are that the in-cloud scavenging should be viewed as a multi-step process, and that a single attachment or removal rate constant does not exist. The concept of the distinction between attachment of particles to cloud and/or raindrops and removal of these particles by rain is introduced in this model. The physical makeup of this in-cloud scavenging scheme together with the size-dependent and time-dependent rate constants are significant contributions for a more adequate understanding of precipitation scavenging than has been achieved heretofore.

From the preliminary results of the present computations in Chapter 3, we may draw the conclusion that the same physical processes are responsible for producing the "inverse" and "direct" relationship. But the proposed model should be viewed only as constructing the conceptual framework for future investigation.

APPENDIX A

TABLE A.

Collision efficiencies for drops of radius R colliding with droplets of radius r at 0° C and 900 mb

$R(\mu m)$	r(μm) 2	3	4	6	8	10	15	20
15		0·003	0·004	0·006	0·010	0·012	0·007	ν —
20	0·002	0·002	0·004	0·007	0·015	0·023	0·026	—
25	—	—	—	0·010	0·026	0·054	0·130	0·06
30	†	†	†	0·016	0·058	0·17	0·485	0·54
40	†	†	—	0·19	0·35	0·45	0·60	0·65
50	†	†	0·05	0·22	0·42	0·56	0·73	0·80
80	—	—	0·18	0·35	0·50	0·62	0·78	0·85
100	0·03	0·07	0·17	0·41	0·58	0·69	0·82	0·88
150	0·07	0·13	0·27	0·48	0·65	0·73	0·84	0·91
200	0·10	0·20	0·34	0·58	0·70	0·78	0·88	0·92
300	0·15	0·31	0·44	0·65	0·75	0·83	0·96	0·91
400	0·17	0·37	0·50	0·70	0·81	0·87	0·93	0·96
600	0·17	0·40	0·54	0·72	0·83	0·88	0·94	0·98
1000	0·15	0·37	0·52	0·74	0·82	0·88	0·94	0·98
1400	0·11	0·34	0·49	0·71	0·83	0·88	0·94	0·95
1800	0·08	0·29	0·45	0·68	0·80	0·86	0·96	0·94
2400	0·04	0·22	0·39	0·62	0·75	0·83	0·92	0·96
3000	0·02	0·16	0·33	0·55	0·71	0·81	0·90	0·94

The removal of the particles and droplets by the raindrop population is based on collision efficiency (E(r,r)) values adopted from Mason (1971, Table A). For particles smaller than 2 μm radius, zero efficiency has been used.

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