

An Analysis of In-Cloud Scavenging

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ABSTRACT

The fate of airborne contaminants that enter a cloud with an overriding rain, independently generated, is considered by means of three differential equations which express the overall mass conservation of contaminant. The model incorporates the physical processes of diffusive attachment, impact collection and accretion, and includes consideration of particle and cloud droplet size spectra. The rainout ratio and rainout efficiency are evaluated on the basis of the theoretical results, and the general expressions for different cloud conditions are proposed.

1. Introduction

The scavenging of contaminant by precipitation is frequently treated in two parts, i.e., that occurring below cloud (washout) and that within cloud (rainout). For the below-cloud case, 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 uncertainty for quantitative prediction of phoretic and electrical influences on the attachment rate, these processes contributing to the total in-cloud scavenging effect are ignored.

2. 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:

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

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

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

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 ($i=1, \dots, 30$)
- 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 giving greater accuracy in the calculations, and 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 *i*th 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 formula

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

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

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

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

$$\frac{dNr_i}{dt} = \bar{\lambda} Nc_i + \sum_{k=1}^N (\beta_{ik} + \gamma_{ik}) Na_i. \tag{6}$$

A physical interpretation of the simplified Eqs. (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 cannot be distinguished, and the number of particles of each size removed by *k*th class raindrops and attached to *j*th class cloud droplets cannot 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.

3. 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, Eqs. (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 as

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

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

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

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

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

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

$$Nc(t) = [Nc(0) + Na(0)(1 - e^{-\alpha t})] e^{-\bar{\lambda} t} + \frac{\alpha Na(0) e^{-\alpha t}}{\bar{\lambda} - (\alpha + \beta + \gamma)} [e^{-(\alpha + \beta + \gamma)t} - e^{-\bar{\lambda} t}], \tag{10}$$

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

$$Nr(t) = [Nc(0) + Na(0)](1 - e^{-\bar{\lambda} t}) + \frac{Na(0) e^{-\alpha t}}{\bar{\lambda} - (\alpha + \beta + \gamma)} [e^{-\bar{\lambda} t} - e^{-(\alpha + \beta + \gamma)t}] [\bar{\lambda} - (\beta + \gamma)]. \tag{11}$$

Eqs. (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

$$\alpha_B = \sum \frac{kT}{3\eta} \left(\frac{1 + al/r_p}{r_p} + \frac{1 + al/r}{r} \right) (r_p + r) M_c, \tag{12}$$

for each *i* and *j*,

where r_p and *r* are particle and droplet radii of each size class, respectively; *a* the Cunningham correction factor (=0.9); *l* the mean free path of air molecules; M_c the number density of cloud droplets of each size class

(cm^{-3}); k Boltzmann's constant; T the absolute temperature; and η 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:

$$\alpha_T = \sum 14.1(r_p+r)^3 \left(\frac{\epsilon}{\nu}\right)^{\frac{1}{2}} M_c, \text{ for each } i \text{ and } j, \quad (13)$$

for the contribution α_T of air turbulence to the attachment rate. Here ϵ is the rate of energy dissipation and ν the kinematic viscosity. The quantity ϵ is dependent upon the nature of the cloud considered. Ackerman (1968) gives a value for ϵ (for the inertial subrange) of $6.4 \text{ cm}^2 \text{ sec}^{-3}$ as the average for stratiform cloud. Since we consider the turbulent and Brownian contributions to be additive,

$$\alpha = \alpha_B + \alpha_T. \quad (14)$$

To complete the evaluation of the diffusive attachment rate, the size spectra of the contaminant particles and the cloud droplets are required. For the latter, Khrgian and Mazin (1952) give

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

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

$$\left. \begin{aligned} a &= (1.45\bar{r}^{-6})Q/\rho_l \\ b &= 3\bar{r}^{-1} \end{aligned} \right\}$$

where Q is the liquid water content (gm cm^{-3}), ρ_l the water density, and $\bar{r} = (1/N) \int_0^\infty rn(r)dr$ 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 ($=2.5$). For the present model we have set $r_m = 10^{-5} \text{ cm}$ 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,

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

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

The same treatment is used to evaluate the diffusive attachment between particles and raindrops. Here, the Marshall-Palmer (1948) raindrop-size formulation is used.

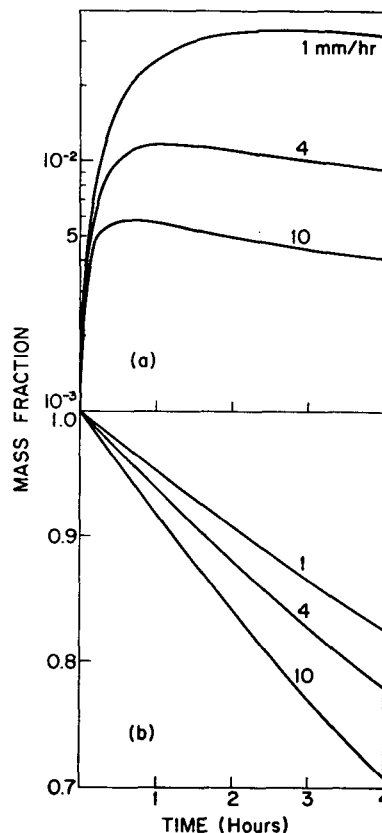


FIG. 1. Case I. Cloud water constant (replenished): (a) fraction of mass attached to cloud droplets and (b) fraction of mass remaining in cloud air as a function of time.

b. Impact collection

The impact collection rate is

$$\gamma = \sum [\pi R^2 E(r,R)(V_R - V_r)N_R \Delta R], \quad (17)$$

for each i and k ,

where V_R is the terminal velocity of raindrops of radius R (Dingle and Lee, 1972), V_r the terminal velocity of particles of radius r , $E(R,r)$ the collision efficiency, and $N_R \Delta R$ the number of raindrops per unit volume in the size range R to $R+\Delta R$. Values of $E(R,r)$ are derived from the table compiled by Mason (1971) after graphic smoothing (Storebø and Dingle, 1974).

c. Accretion

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

4. Results

Four cases have been selected to bring out the scavenging effects of various parameters of the model. In the computations, the cloud spectrum and size distribution of pollution are assumed to be time-

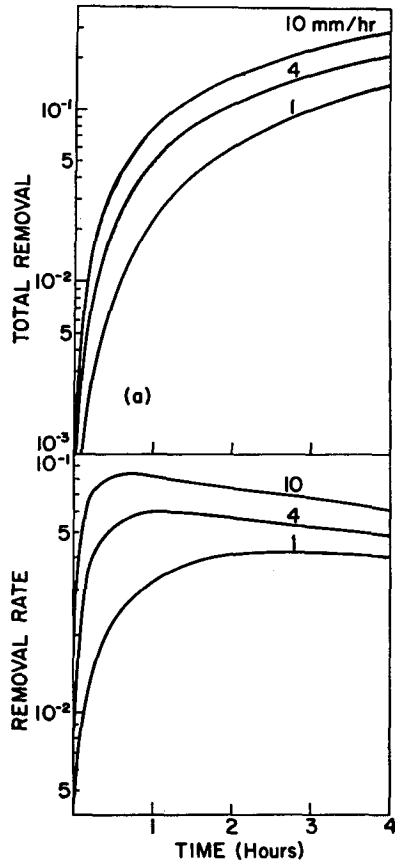


FIG. 2. Case I. Cloud water constant (replenished): (a) integral fraction of mass removed by rainfall and (b) removal rate (mass fraction hr^{-1}) as a function of time.

independent for a single time step, and the term $N_c(0)$ of Eqs. (8), (10) and (11) is not accounted for. To simplify our computations, we assume that the parameters $\alpha, \beta, \gamma, \bar{\lambda}$ 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} to 10^{-3} sec^{-1} . For rainfall rates $< 4 \text{ mm hr}^{-1}$, the cloud water removed every 2 min does not significantly change the cloud droplet concentration. Therefore, the parameters may be considered constant for this time interval. For rainfall rates $> 4 \text{ mm hr}^{-1}$, the time step is reduced to 1 min and the parameters are readjusted accordingly.

a. Case I

Steady-state cloud: Cloud water is replenished at a rate exactly equal to the rate of removal of cloud water by falling rain. Removal of pollution particles is by 1) Brownian and turbulent attachment to cloud droplets and falling rain, and 2) removal of cloud water with attached pollution by falling rain. Results for this case are shown in Figs. 1 and 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 $< 2 \mu\text{m}$ radius are not captured by raindrops [i.e., $E(R,r) \rightarrow 0$] and particles $> 2 \mu$ 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. It is found that about 29% of the contaminant is scavenged by a rainfall of intensity 10 mm hr^{-1} in 4 hr. The removal rate (mass fraction hr^{-1}) 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 Figs. 1b and 3b

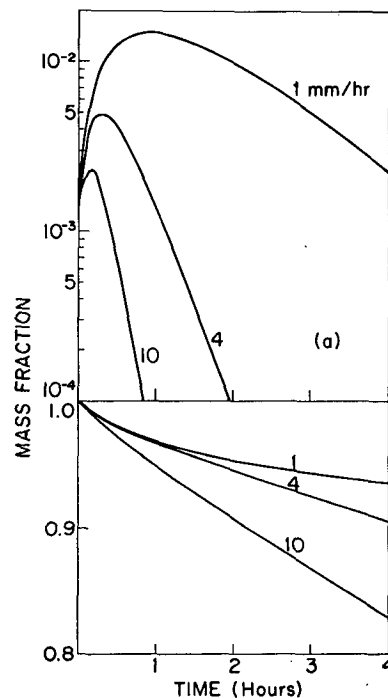


FIG. 3. Case II. Decreasing cloud water: (a) fraction of mass attached to cloud droplets and (b) fraction of mass remaining in cloud air as a function of time.

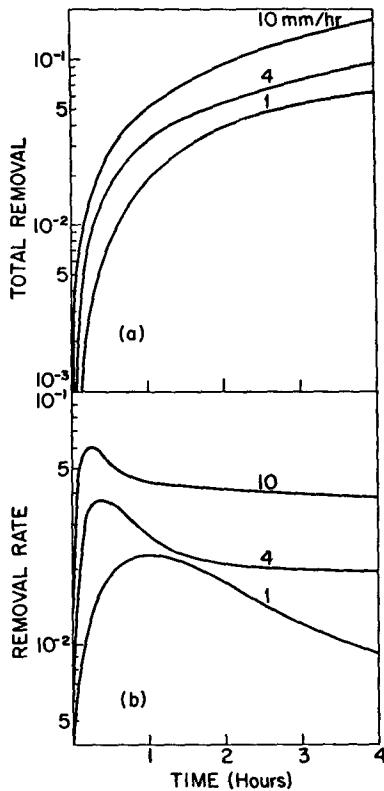


FIG. 4. Case II. Decreasing cloud water: (a) integral fraction of mass removed by rainfall and (b) removal rate (mass fraction hr⁻¹) as a function of time.

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. Cases III and IV

Case III is similar to Case I except that a 2-hr period of resident of pollution particles in the cloud is allowed prior to the onset of rain. Case IV is similar to Case III except that the cloud is non-steady as in Case II.

In these cases, the in-cloud mixing time t_c is 2 hr, and rain falls through the polluted cloud for another 2 hr. 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 2 hr during which the rainfall rate is 10 mm hr⁻¹, 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 2 hr.

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

$$K = \chi_0 \frac{\rho_l}{Q} E, \tag{18}$$

where K is the concentration of a specified contaminant in rainwater, χ_0 the concentration of the same contaminant originally in the air, and E the rainout

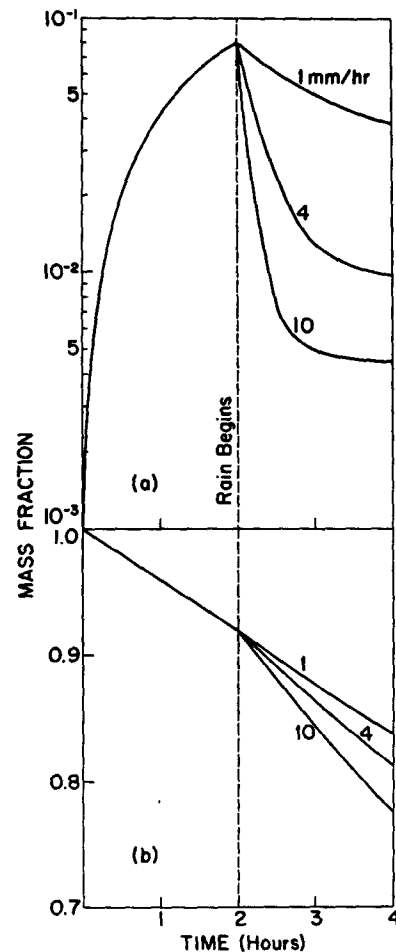


FIG. 5. Case III. Cloud water constant (replenished) with rain beginning at time $t=2$ hr: (a) fraction of mass attached to cloud droplets and (b) fraction of mass remaining in cloud air as a function of time.

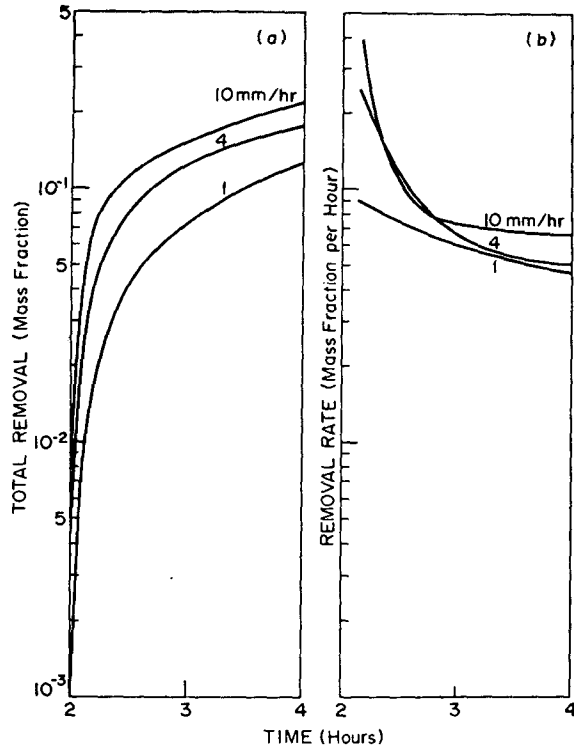


FIG. 6. Case III. Cloud water constant (replenished) with rain beginning at time $t=2$ hr (see Fig. 5): (a) integral fraction of mass removed by rainfall and (b) removal rate as a function of time.

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:

$$F_1 = 1 - (0.044 + 0.0042I)t^{0.9}, \quad \text{(steady state, case I)} \quad (19a)$$

$$F_2 = 1 - (0.028 + 0.0017I^{1.35})t^{0.68}, \quad \text{(non-steady state, case II)} \quad (19b)$$

$$F_3 = 1 - [(0.044 + 0.0042I)t^{0.90} + 0.08], \quad \text{(steady state, case III)} \quad (19c)$$

$$F_4 = 1 - [(0.028 + 0.0017I^{1.35})t^{0.68} + 0.08], \quad \text{(non-steady state, case IV)} \quad (19d)$$

where F is the mass fraction remaining in cloud air, I the rainfall intensity (mm hr^{-1}), and t the precipitation time (hr). With these equations, the rainout efficiency for each case can be expressed as

$$E_1 = 1 - F_1 = (0.044 + 0.0042I)t^{0.90}, \quad (20a)$$

$$E_2 = 1 - F_2 = (0.028 + 0.0017I^{1.35})t^{0.68}, \quad (20b)$$

$$E_3 = 1 - F_3 = (0.044 + 0.0042I)t^{0.90} + 0.08, \quad (20c)$$

$$E_4 = 1 - F_4 = (0.028 + 0.0017I^{1.35})t^{0.68} + 0.08. \quad (20d)$$

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

$$\frac{K}{\chi_0} = E_i \frac{\rho_l}{Q}. \quad (21)$$

This shows that the rainout ratio varies directly with precipitation rate and duration of rainfall. With reasonable values of Q (here $Q = 3.0 \times 10^{-7} \text{ gm cm}^{-3}$), the rainout efficiency and the rainout ratios can be determined. Figs. 9 and 10 show these results which compare favorably with experimental observations (see Table 1).

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

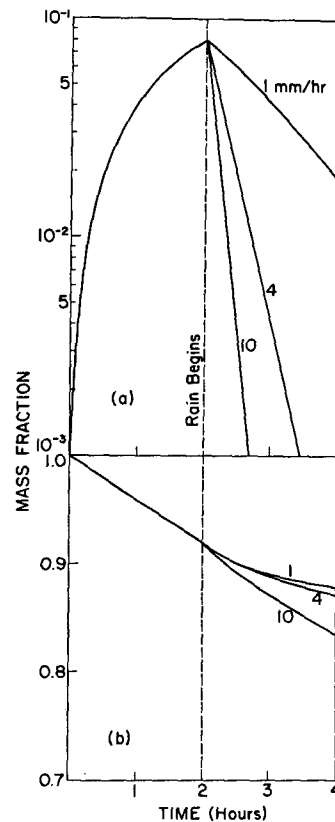


FIG. 7. Case IV. Decreasing cloud water with rain beginning at time $t=2$ hr: (a) fraction of mass attached to cloud droplets and (b) fraction of mass remaining in cloud air as a function of time.

TABLE 1. 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 ⁻¹) | | | | |
| 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 ⁻¹) | | | | |
| 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 |

* Small, 1960; Hinzpeter, 1958.

** Water equivalent.

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

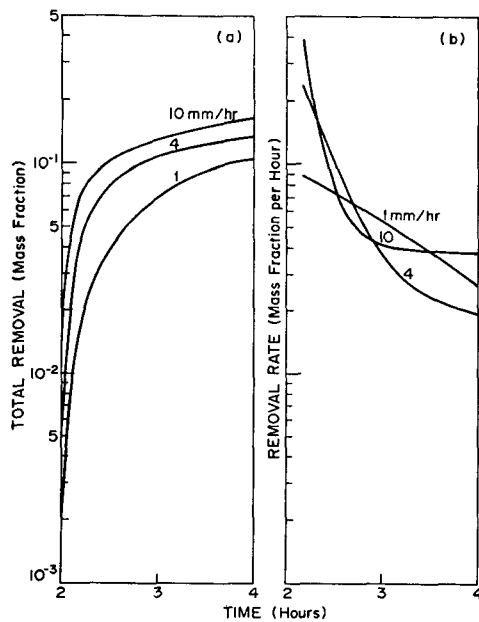


FIG. 8. Case IV. Decreasing cloud water with rain beginning at time $t = 2$ hr (see Fig. 7): (a) integral fraction of mass removed by rainfall and (b) removal rate as a function to time.

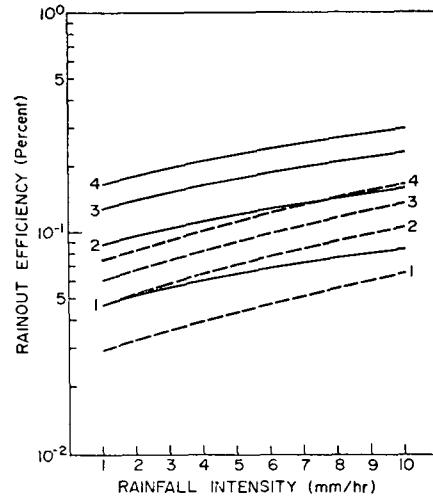


FIG. 9. Rainout efficiency vs rainfall intensity for Case I (solid) and Case II (dashed). Precipitation time in hours is indicated by the numbers.

of in-cloud scavenging wherever observations or data sources are inadequate.

5. Conclusion

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 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

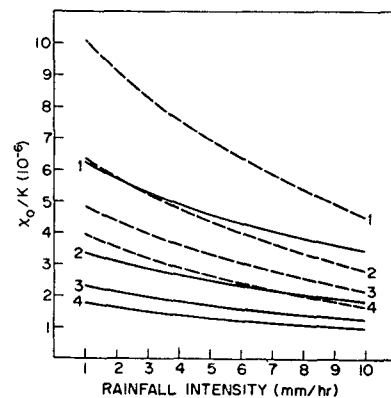


FIG. 10. χ_0/K vs rainfall intensity for Case I (solid) and Case II (dashed). Precipitation time in hours is indicated by the numbers.

(Cases I and 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 and 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 new insights into the influence of such parameters as size spectrum, cloud water content, rainfall rate, and attachment mechanism in the scavenging effect.

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. Although our model includes what we feel to be the significant in-cloud scavenging processes, it is recognized that electrical charge distributions in clouds are not well understood, that the rates of energy dissipation by relevant frequency bands in the turbulence spectrum are inadequately known, and that phoretic processes contributing to in-cloud scavenging remain somewhat uncertain. However, we propose here a model that appears to be of general validity and one which will continue to hold when these additional physical inputs to the system are more adequately treated.

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