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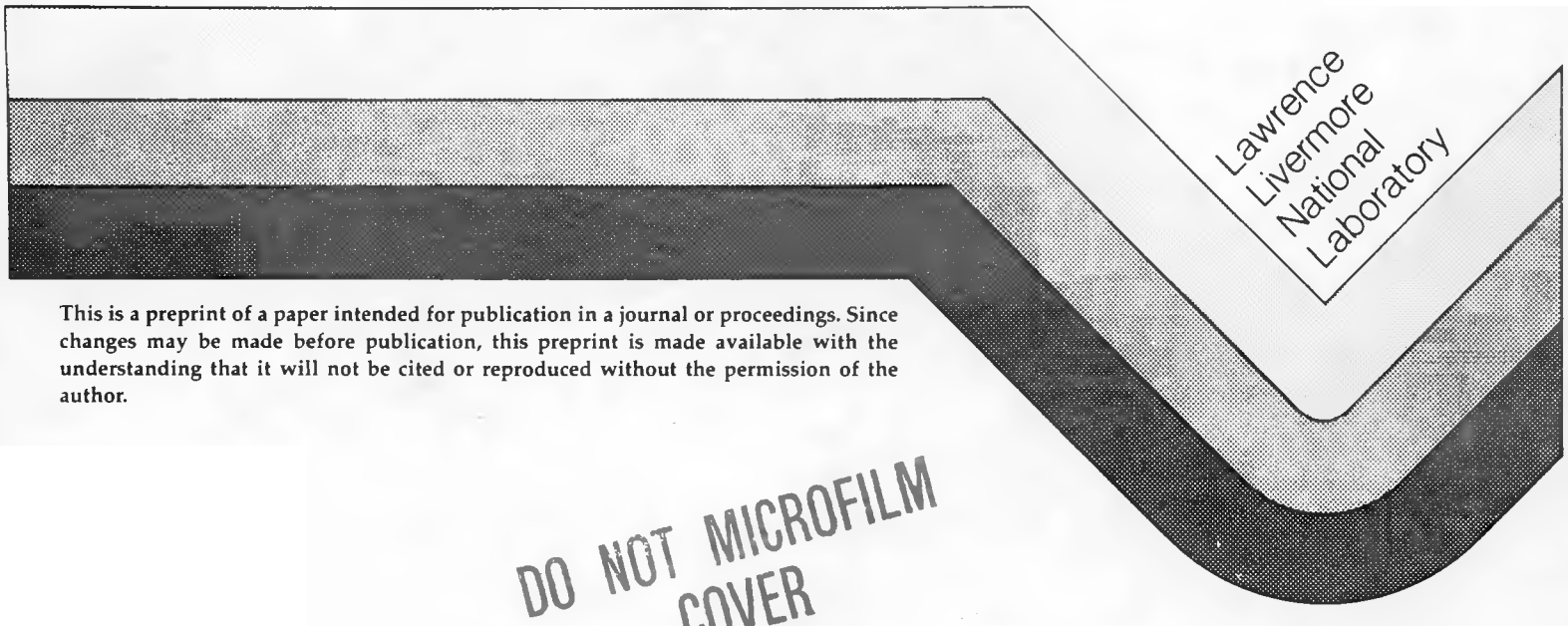

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PARAMETERIZATION OF AEROSOL SCAVENGING IN A CONVECTIVE CLOUD MODEL*

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1. INTRODUCTION

A three-dimensional numerical cloud model (OCTET) is being developed to simulate the transport, diffusion and scavenging of aerosol particles. The initial purpose of the model is to estimate the fraction of smoke particles emitted by large fires that are scavenged by precipitation immediately above the fire and the fraction that is injected into the upper troposphere and stratosphere. Because large fires generate strong updrafts, the clouds penetrate above the freezing level and contain frozen hydrometeors; therefore, interactions of the aerosol with the ice phase must be included in the model. This paper is a description of the current microphysics and scavenging parameterization.

2. DYNAMICS

The OCTET modeling system simulates the three-dimensional, moist, nonhydrostatic circulations of natural and fire-driven convective storms. It is being used to simulate the dynamics, microphysics and scavenging of natural precipitating clouds and large city fires with energy release rates on the order of 10-100 kw/m^2 over urban areas of tens to hundreds of square kilometers. The original OCTET model was based on the Klemp and Wilhelmson (1978) storm model; additional information on the OCTET modeling system is given elsewhere in this preprint volume (Bradley and Molenkamp, 1990).

3. CLOUD MICROPHYSICS

The representation of cloud microphysics is a bulk-water parameterization. A parallel parameterization represents the scavenging interactions between pollutant particles and hydrometeors including nucleation, attachment, deposition and resuspension as well as the transfer interactions as particles collected by one type of hydrometeor are transferred to another type during freezing, melting, coagulation, etc. of the hydrometeors.

The development of the cloud microphysics parameterization started from a version of the Orville parameterization (Lin, Farley and Orville, 1983, referred to as LFO below). It includes water in six forms (vapor, cloud droplets, rain, ice crystals, snow, and graupel), and allows for transfer of water between phases because of condensation, evaporation, sublimation, deposition, autoconversion, accretion, freezing, melting, shedding, and riming. The rain drops, snow and graupel are assumed to fall relative to the updraft while the cloud droplets, ice crystals and water vapor have negligible terminal velocities. Figure 1 shows the six water phases and the transfer processes currently included in the model. Figure 2 shows the corresponding transfers of aerosol particles between and within the various water phases. Except for those processes described below the transfer rates are those given by LFO.

3.1 Saturation Adjustment

The condensation and evaporation of cloud droplets is not treated with a transfer rate but as an isobaric adjustment, since supersaturation with respect to liquid is always very small in the atmosphere. For temperatures below freezing, both cloud water and ice crystals may be present, and it is not completely clear how to perform the saturation adjustment. Orville assumes that the adjusted saturation vapor pressure is a weighted average of the saturation vapor pressures with respect to water and ice; the weighting is based on the amount of cloud water and cloud ice present. We perform the saturation adjustment with respect to liquid only, changing the cloud water mixing ratio to be at or below water saturation at all temperatures. This produces an equilibrium state for the liquid and leaves the cloud ice in a supersaturated state, but that seems to be a better approach since ice crystals do not take up vapor rapidly enough to stay in saturation balance.

This process is important for scavenging since the Bergeron process, in which ice crystals grow by deposition while the cloud water evaporates, can lead to resuspension of particles collected by cloud droplets without transferring them to the ice crystals. The assumption that the saturation adjustment only applies to water saturation permits evaluation of the diffusional growth of ice crystals as a rate process rather than an adjustment.

3.2 Ice Crystal Growth by Deposition and Riming

With the saturation adjustment being performed with respect to liquid only, the formation of ice crystals depends on ice nucleation and the freezing of cloud droplets while their growth depends explicitly on vapor deposition and riming. As they grow ice crystals can become large enough to be considered snow. These effects are accounted for by five new rates that replace the three terms, P_{IDW}, P_{SFW}, and P_{SFI} in LFO. These new rates are P_{IDV},

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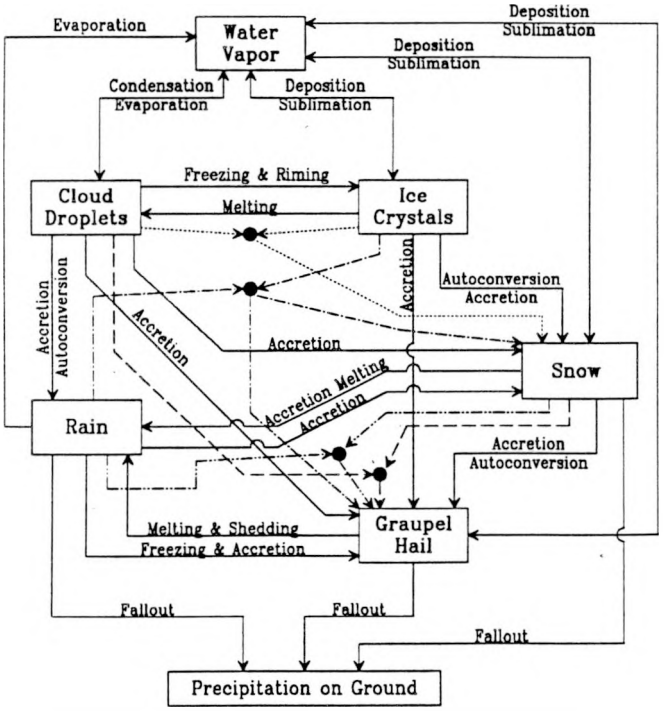

Fig. 1. Cloud microphysical processes. Three-way processes, indicated by non-solid arrows passing through small circles, represent the coagulation of two types of hydrometeors followed by freezing to form a third type.

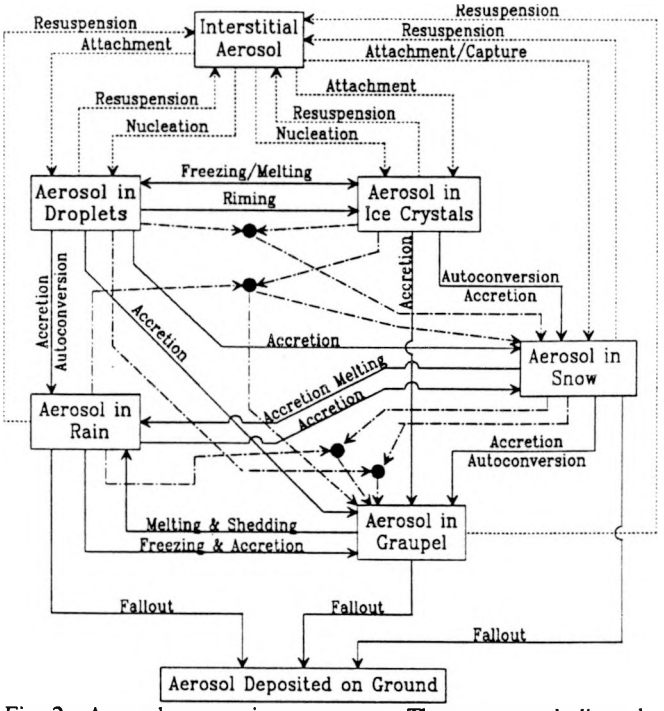

Fig. 2. Aerosol scavenging processes. The processes indicated with solid arrows and the three-way processes are assumed to occur at rates which are proportional to the corresponding water transfer rates.

the depositional growth of ice crystals, P_{SDV}, the depositional growth of ice crystals that grow into snowflakes, P_{IACW}, the riming growth of ice crystals, P_{SRW}, the riming growth of ice crystals that become snowflakes and P_{SRDI}, the cloud ice that becomes snow due to deposition and riming.

Deposition of vapor on cloud ice is limited not by the supersaturation with respect to ice but by the rate of growth of ice crystals. As in LFO, the growth rate of an ice crystal of mass m_i at water saturation is assumed to be

$$\frac{dm_i}{dt} = a_1 m_i^{a_2} \tag{1}$$

where a_1 and a_2 are constants derived from observations and tabulated over a temperature range from 0 to -31 C (Koenig, 1971). For temperatures below -40 C, the values for a_1 and a_2 have been set to 5×10^{-5} and 0.5923 to represent more rapid diffusional growth at cold temperatures. Between -31 and -40 C, the constants are interpolated at integer temperatures in a manner consistent with the values in Koenig's table. These assumptions have been made since there does not seem to be any data on ice crystal growth rates at such low temperatures. In fact, the only recent data on depositional growth of ice crystals seems to be the work of Fukuta (summarized in Redder and Fukuta, 1989) which applies for temperatures warmer than -20 C. The ice crystal mass used in (1) is the mean mass of an ice crystal,

$$m_i = \frac{\rho_a q_i}{n_I}, \tag{2}$$

where ρ_a is the air density, q_i the ice water mixing ratio and n_I the ice crystal number concentration (Fletcher, 1962) which is

$$n_I = n_{0I}\exp(-\beta T_c). \tag{3}$$

Here T_c is the temperature in deg C and n_{0I} and β are constants with values of 10^{-5} m^{-4} and 0.5 deg^{-1}. During the depositional growth of ice crystals the atmospheric vapor pressure is between water and ice saturation; therefore, the mass growth rate is multiplied by a saturation fraction, s_f, where

$$s_f = \frac{q_v - q_{i,s}}{q_{w,s} - q_{i,s}}. \tag{4}$$

The rate of riming of an ice crystal of radius r_i is given by

$$\frac{dm_i}{dt} = \pi E_{IW} r_i^2 V_i(r)\rho_a q_c \tag{5}$$

where E_{IW} is the collection efficiency of ice crystals for cloud water, q_c the cloud water mixing ratio and V_i the terminal velocity of an ice crystal, which is given by

$$V_i(r_i) = a_I r_i / r_t \tag{6}$$

where a_I is 1 m/s and r_t is 50 μm.

As in LFO, an ice crystal that becomes larger than 50 μm is assumed to move into the snow category. The sum of the ice crystal depositional and riming growth rates, (1) and (5), can be integrated over a model time step to determine a critical size, r_c, for which an ice crystal will just grow to the maximum ice crystal size of 50 μm. This size is given by

$$r_c = \left(\frac{AF}{1-BF}\right)^{\frac{1}{d}} \tag{7}$$

where

$$A = \frac{E_{IW} a_1 \rho_a q_c}{4 \rho_I r_t}, \tag{8}$$

$$B = \frac{a_1 s_f (4\pi\rho_I)^{a_2-1}}{3^{a_2}}, \tag{9}$$

$$d = 3(a_2-1) \tag{10}$$

and

$$F = \exp\left[\ln\left(\frac{r_t}{A+Br_t^d}\right)-Ad\Delta t\right]. \tag{11}$$

In these expressions ρ_I is the density of ice crystals and Δt is the model time step.

Once r_c has been determined, the five production rates can be estimated by integrating the size dependent growth rates (1) and (5) over an assumed ice crystal size distribution. Comparison of the transfer from ice to snow for a number of possible size distributions revealed surprisingly little dependence on the assumed size distribution; therefore, a simple distribution spreading mass uniformly over an interval from r_b to r_t is used,

$$n_I(r) = \begin{cases} 0 & r \le r_b ; r \ge r_t \\ n_{0I} r^{-3} & r_b < r < r_t \end{cases} \tag{12}$$

where

$$n_{0I} = \frac{3\rho_a q_I}{4\pi\rho_I (r_t - r_b)}. \tag{13}$$

Using this size distribution

$$P_{IDV} = s_f a_1 n_{0I} \left(\frac{4}{3}\pi\rho_I\right)^{a_2} \left[\frac{r_c^{3a_2-2} - r_b^{3a_2-2}}{3a_2-2}\right], \tag{14}$$

$$P_{SDV} = s_f a_1 n_{0I} \left(\frac{4}{3}\pi\rho_I\right)^{a_2} \left[\frac{r_t^{3a_2-2} - r_c^{3a_2-2}}{3a_2-2}\right], \tag{15}$$

$$P_{IACW} = \frac{\pi E_{IW} \rho_a q_c n_{0I} a_1 (r_c - r_b)}{r_t}, \tag{16}$$

$$P_{IACW} = \frac{\pi E_{IW} \rho_a q_c n_{0I} a_1 (r_t - r_c)}{r_t} \tag{17}$$

and

$$P_{SRDI} = \frac{r_t - r_c}{r_t - r_b} \rho_a q_I. \tag{18}$$

3.3 Sublimation of Ice

The sublimation of ice when the atmosphere is subsaturated with respect to ice is evaluated by multiplying the rate of loss of mass for an ice crystal with the mass mean radius, r_i, by the number of ice crystals. The rate of mass loss is given by Pruppacher and Klett (1978),

$$\frac{dm_i}{dt} = \frac{4\pi(S_i-1)r_i}{A_i+B_i}, \tag{19}$$

where S_i is the saturation ratio with respect to ice ($S_i < 1$),

$$A_i = \frac{L_s^2}{k_a R_w T^2} \quad \text{and} \quad B_i = \frac{1}{\rho_a q_{i,s} \psi}. \tag{20}$$

Here L_s is the latent heat of sublimation, R_w the specific gas constant for water vapor, k_a the thermal conductivity of air, ψ the diffusivity of water vapor, and $q_{i,s}$ the saturation mixing ratio with respect to ice.

The number of ice crystals n_I is given by (3) so that

$$P_{ISUB} = \frac{n_I}{\rho_a} \frac{dm_i}{dt}. \tag{21}$$

3.4 Cloud Droplet Freezing

With saturation adjustment evaluated with respect to liquid at all temperatures, it is possible for cloud water to form at temperatures well below -40 C. Homogeneous freezing of cloud droplets occurs at these low temperatures, but rather than treat -40 C as a threshold for homogeneous freezing of cloud droplets, a mean droplet lifetime, $L(T)$, is defined such that the mean life of a droplet at -40 C is 1 min and decreases by an order of magnitude for every 4 deg of additional cooling,

$$L(T_c) = \frac{60}{10^{\left(\frac{T_f - T_c}{4}\right)}}. \tag{22}$$

Then the rate of homogeneous freezing of cloud water is

$$P_{IHFW} = \frac{q_c}{L(T_c)}. \tag{23}$$

4. SCAVENGING MICROPHYSICS

The primary mechanism for removing aerosol particles from the atmosphere is precipitation scavenging. Aerosol particles, incorporated into hydrometeors either as condensation or deposition nuclei or by attachment, move with the hydrometeors until the particles are deposited on the ground or resuspended when the hydrometeor evaporates completely. A fraction of the aerosol ingested into the cloud is removed and the rest is redistributed in the atmosphere with modified properties. The aerosol may also have significant effects on the microphysical development of the cloud, but these effects have not yet been included in the model.

In the bulk-water representation of scavenging, aerosol particles can be associated with each phase of hydrometeor (cloud droplets, ice crystals, rain, snow and graupel). Aerosol particles not associated with hydrometeors are termed dry aerosol even if they have acquired some moisture because of their solubility. The aerosol associated with each phase and the interaction processes

being incorporated into the model are shown in Figure 2. The scavenging parameterization is incomplete; therefore, expressions for some of the interaction rates have not yet been determined.

Assuming that the aerosol contained in the various types of hydrometeors is well-mixed, the rate of transfer of aerosol between phases because of accretion, autoconversion, melting, freezing, and riming is proportional to the corresponding rate of transfer of water. These transfers are indicated by solid lines in Figure 2. The three-way processes, indicated in Figure 2 by dot-dashed lines, are essentially accretion processes that have aerosol transfer rates proportional to the water transfer rates.

4.1 Condensation Nucleation Scavenging

The representation of condensation nucleation is based on results from a detailed parcel model (CAMP) that estimates the fraction of aerosol particles activated as condensation nuclei by calculating the evolution of the aerosol and droplet size distributions (Edwards and Penner, 1988; Edwards, 1989). Using results from many CAMP simulations over the appropriate range of updraft speeds and aerosol characteristics, a table of the fraction nucleated as a function of updraft is constructed. Whenever aerosol is ingested through any cloud boundary where there is an upward component of motion this table is used to specify the mass fraction that is incorporated into cloud droplets. No additional aerosol is scavenged in the interior of the cloud through condensation nucleation since all subsequent condensation of water is assumed to occur on the droplets that are already present.

4.2 Wet Hail Growth and Shedding

Falling graupel can rapidly accrete large quantities of cloud and rain water, and, when the temperature is near 0 C, some of this collected liquid does not freeze but is shed as rain. Since the accreted cloud and rain water contains pollutant, the water that is shed also contains pollutant. The assumption is that the fraction of accreted pollutant shed is the same as the fraction of collected water shed.

4.3 Resuspension by Evaporating Cloud Droplets

The evaporation of cloud droplets occurs for two reasons, 1) droplets are transported into regions of subsaturation and 2) ice crystals form and grow by deposition inside the cloud tending to reduce the vapor pressure which leads to droplet evaporation. When a cloud droplet evaporates completely, its collected aerosol is resuspended as a dry particle. The earlier assumption that the rate of resuspension is proportional to the rate of cloud water evaporation is an over-estimate of the resuspension rate (Molenkamp, 1974).

The fraction of the aerosol in cloud water that is resuspended because of droplet evaporation is equal to the fraction of the cloud water in droplets that evaporate completely during a time step. If the size of the largest droplet that evaporates completely in a time step is r_s, then the initial mass of water in drops that evaporate completely is

$$M_{EC} = \frac{4}{3}\pi\rho_w \int_0^{r_s} r^3 n(r)\,dr \qquad (24)$$

where ρ_w is the density of liquid water and $n(r)$ is the cloud droplet size distribution. r_s can be evaluated if it is assumed that the subsaturation, S, is constant over a time step by integrating the diffusional growth equation,

$$r\frac{dr}{dt} = \frac{(S-1)}{\rho_w[A_w+B_w]} = \frac{(S-1)}{G}, \qquad (25)$$

over time. In (25)

$$A_w = \frac{L_c^2}{k_a R_w T^2}, \quad \text{and} \quad B_w = \frac{1}{\rho_a q_{w,s}\psi}. \qquad (26)$$

with L_c the latent heat of condensation, and $q_{w,s}$ the saturation mixing ratio with respect to liquid. A_w, B_w and therefore G are constants at any location in the model. Integration of (25) with respect to time gives

$$r(t) = \left[r^2 + \frac{2(S-1)}{G}t\right]^{1/2}. \qquad (27)$$

From (27) r_s, the radius of the drop that just evaporates in time Δt, can be related to S by

$$r_s^2 = -\frac{2(S-1)}{G}\Delta t. \qquad (28)$$

From the saturation adjustment step, the mass of the cloud water not evaporated, $\rho_a q_{c,a}$ is known and can be set to the initial cloud water amount, $\rho_a q_{c,b}$ minus the mass in drops that evaporate completely and minus the mass lost from drops that partially evaporate,

$$\rho_a q_{c,a} = \rho_a q_{c,b} - \int_0^{r_s} m(r)n(r)\,dr + \int_{r_s}^{\infty} \Delta m(r)n(r)\,dr, \qquad (29)$$

where $m(r)$ is the mass of a droplet of radius r and $\Delta m(r)$ is the change in mass of a droplet of initial size r during Δt assuming S is constant. $\Delta m(r)$ can be evaluated using (27) and integrating over a time step,

$$\Delta m(r) = \frac{4\pi\rho_w}{3}\left\{\left[r^2+\frac{2(S-1)}{G}\Delta t\right]^{3/2} - r^3\right\}. \qquad (30)$$

Substituting in (29) and using (28) to eliminate the dependence on the unknown saturation ratio

$$\rho_a q_{c,a} = \frac{4\pi\rho_w}{3}\int_{r_s}^{\infty} (r^2 - r_s^2)^{3/2} n(r)\,dr. \qquad (31)$$

The Khrgian-Mazin size distribution (Pruppacher and Klett, 1978) is assumed for cloud droplets,

$$n(r) = \frac{\gamma^3 N_T}{2} r^2 \exp(-\gamma r), \qquad (32)$$

where N_T is the total number of cloud drops and γ can be related to q_c or the mass mean drop size. (24) can be rewritten as the mass fraction of cloud water in droplets that do not completely evaporate,

$$f_{NEC} = e^{-\gamma r_s}\sum_{k=0}^{5}\frac{(\gamma r_s)^k}{k!}, \qquad (33)$$

where r_s is unknown. Now $1\text{-}f_{NEC}$ is equal to the fraction of the aerosol resuspended. (31) can be changed into the fraction of the

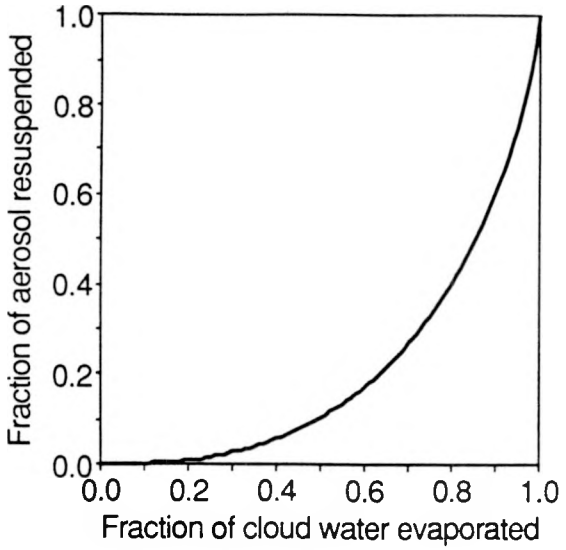

Fig. 3. Fraction of aerosol in cloud water resuspended as a function of cloud water evaporated.

initial cloud water that does not evaporate,

$$f_{NE} = \frac{q_{c,a}}{q_{c,b}} = \frac{\gamma^6}{120}\int_{r_s}^{\infty}[r^2 - r_s^2]^{\frac{3}{2}} r^2 \exp(-\gamma r)dr, \qquad (34)$$

and this fraction is known from the saturation adjustment. Unfortunately this expression can not be integrated analytically to provide an equation in r_s, but (34) can be evaluated numerically for a set of r_s values and used with (33) evaluated at the corresponding values of r_s to produce a table that relates the fraction of aerosol resuspended to the fraction of cloud water evaporated. This relationship is shown in Figure 3.

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