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A NUMERICAL MODEL OF AEROSOL SCAVENGING, PART I: MICROPHYSICS PARAMETERIZATION*

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ABSTRACT

We have developed a three-dimensional numerical model (OCTET) to simulate the dynamics and microphysics of clouds and the transport, diffusion and precipitation scavenging of aerosol particles. In this paper we describe the cloud microphysics and scavenging parameterizations. The representation of cloud microphysics is a bulk-water parameterization which includes water vapor and five types of hydrometeors (cloud droplets, rain drops, ice crystals, snow, and graupel). A parallel parameterization represents the scavenging interactions between pollutant particles and hydrometeors including collection of particles because of condensation nucleation, Brownian and phoretic attachment, and inertial capture; resuspension because of evaporation and sublimation; and transfer interactions where particles collected by one type of hydrometeor are transferred to another type by freezing, melting, accretion, riming and autoconversion.

1. INTRODUCTION

Precipitation scavenging is the primary mechanism for removing small aerosol particles from the atmosphere. This removal depends on both the dynamic characteristics of clouds and microphysical interactions between particles and hydrometeors. To improve our understanding of precipitation scavenging we have developed a numerical model (OCTET) that interactively simulates cloud dynamics, microphysics and scavenging. This model has also been designed to be used as an aid in understanding processes in individual clouds, thunderstorms and mesoscale convective systems and to investigate the vital role clouds play in global climate through latent heating, precipitation, vertical mixing and cloud-radiative feedback.

The development of this model resulted from our need to estimate the net injection, after prompt scavenging, and vertical distribution of smoke from a large number of massive fires ignited by a hypothetical nuclear exchange. In this paper we provide a brief description of the dynamic and cloud microphysical aspects of the model and a more detailed description of the scavenging components. In our companion paper (Bradley and Molenkamp, 1991) we describe a simulation of a hypothetical large city fire.

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2. OCTET SIMULATION SYSTEM

The OCTET Simulation System has been designed to model convective and stratiform clouds, mesoscale storm systems, smoke plumes, and mesoscale circulations. The name OCTET comes from the design of the simulation system as a hierarchy of eight models as shown if Figure 1. A particular OCTET model is generated from the master source code using preprocessing directives and a conditional compiler. Of the eight OCTET models six have been completed; the two electrified models remain as future projects. Two advantages of the hierarchical structure are that coding common to more than one component need be written only once and there is only one master code to maintain.

The dynamics of the OCTET simulation system are based on a three-dimensional, nonhydrostatic, compressible framework similar to that of Klemp and Wilhelmson (1978) with wave-permeable lateral boundaries and a turbulence parameterization based on a time- and space-dependent turbulent energy equation. The prognostic variables of the dry model are the three velocity components (*u*, *v*, and *w*), pressure perturbation, potential temperature, turbulent kinetic energy and water vapor mixing ratio. As additional microphysical detail is added in higher level models, prognostic equations for the mixing ratios of new classes of hydrometeors are added, and, for the models with scavenging, prognostic equations are added for mixing ratios of aerosol associated with each type of hydrometeor. Besides the hierarchical structure of the model there are options for two different warm cloud parameterizations, Lagrangian tracer/samplers, an interface with the LLNL CAMP detailed microphysics model (Edwards and Penner, 1988; Edwards, 1989; Chuang *et al.*, 1990, Penner *et al.*, 1990) for condensation nucleation scavenging, time-variable local heat and smoke sources for fire simulations, aerosol and water mass budgets, and hierarchical data format (HDF) output files for three-dimensional interactive graphical analysis and visualization post-processors.

	No Aerosol	Aerosol
Dry Model (no condensation)	OCTET/v 7 Prognostic Variables	OCTET/va 8 Prognostic Variables
Warm Cloud Model	OCTET/w 9 Prognostic Variables (Klemp-Wilhelmson Cloud Model)	OCTET/wa 12 Prognostic Variables
Cold Cloud Model	OCTET/c 12 Prognostic Variables (Orville-type Microphysics)	OCTET/ca 18 Prognostic Variables
Electrified Cloud Model	OCTET/e 18 Prognostic Variables 1 Diagnostic Variable	OCTET/ea 25 Prognostic Variables 1 Diagnostic Variable

FIGURE 1. The OCTET Simulation System.

3. CLOUD MICROPHYSICS PARAMETERIZATION

Cloud microphysics is represented by a bulk-water parameterization based on Lin *et al.* (1983). In addition to water vapor, five types of hydrometeors are included: cloud droplets, rain, ice crystals, snow and graupel. There is a prognostic conservation equation for the mass mixing ratio of each type of hydrometeor, q_j,

$$\frac{\partial q_j}{\partial t} = - V\cdot\nabla q_j + \frac{1}{\rho}\frac{\partial}{\partial z}(U_j q_j \rho) + \nabla\cdot K_m \nabla q_j + P_j, \tag{1}$$

where V is the air velocity vector, K_m the eddy diffusion coefficient for momentum, ρ the air density, U_j the mass-weighted terminal velocity for hydrometeor type j, and P_j the net rate of production of hydrometeor type j. The terminal velocities for cloud droplets and ice crystals are assumed to be 0, and the second term on the right is dropped from those equations. The net rate of production is determined by a sum of the sources and sinks for the transfer processes indicated in Figure 2. The individual transfer rates, P_{jppk}, are listed in Table 1. In the subscript, the middle two characters indicate the process, the last

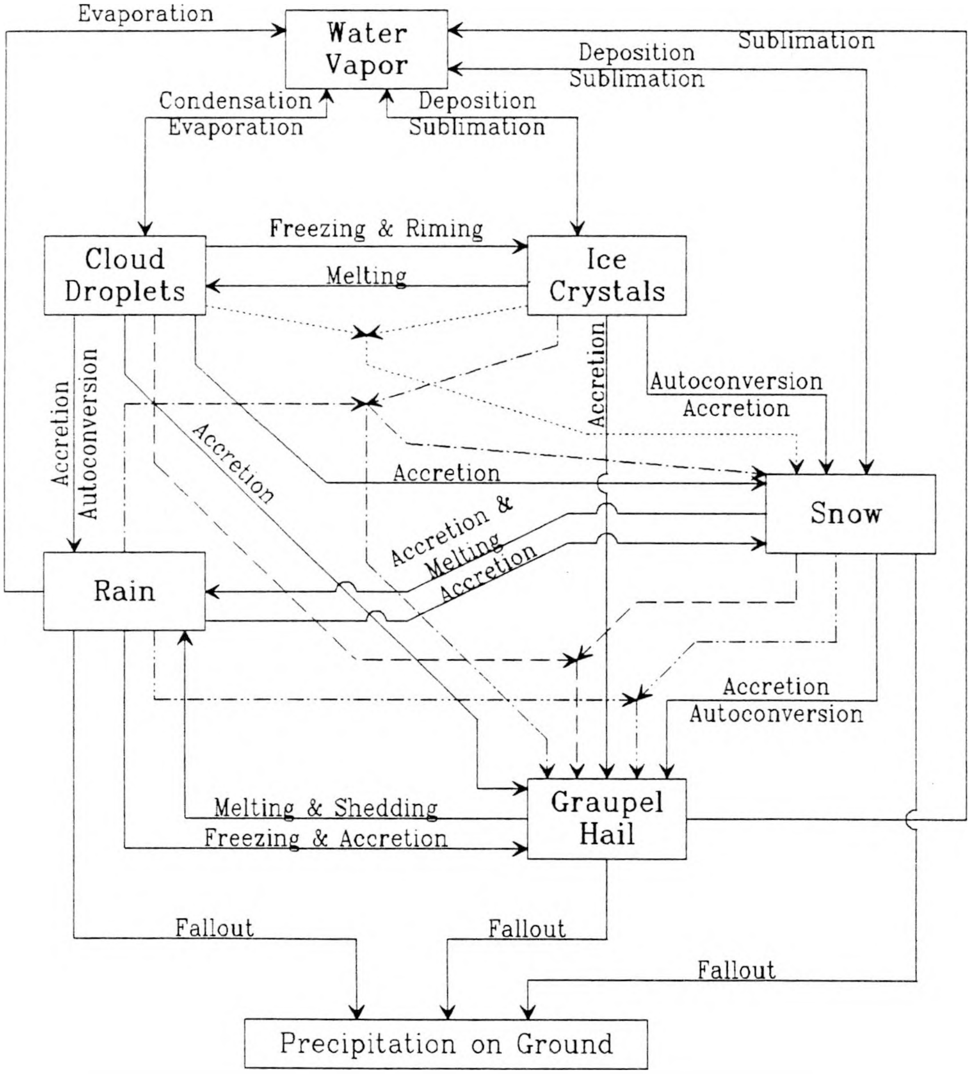

FIGURE 2. Cloud microphysical processes included in parameterization. Processes indicated by two arrows coming together and going to a third type represent coagulation followed by freezing.

character the source, and the first character the sink, although the sink depends on temperature or mixing ratio in some cases.

With just a few important exceptions, we use the transfer rates of Lin, *et al.* (1983). The most significant exceptions are condensation of vapor on droplets where we always

TABLE 1. Microphysics and Scavenging Processes Included in the Parameterization.

Subscript	Sink*	Source*	Proportional Scavenging	Process
CCNV	C	V	No	Condensation
CMLI	C	I	Yes	Melting
GACC	G,R†	C	Yes	Accretion
GACI	G	I	Yes	Accretion
GACR	G	R	Yes	Accretion
GACS	G	S	Yes	Accretion
GAUS	G	S	Yes	Autoconversion
GDPV	G	V	0§	Deposition
GFRR	G	R	Yes	Freezing
GWET	G	C,I,R,S	Yes	Accretion (wet growth)
IACR	S,G	R	Yes	Accretion
IDPV	I	V	0§	Deposition
IHFC	I	C	Yes	Homogeneous freezing
IRIC	I	C	Yes	Accretion (riming)
RACC	R	C	Yes	Accretion
RACI	S,G	I	Yes	Accretion
RACS	G,R†	S	Yes	Accretion
RAUC	R	C	Yes	Autoconversion
RMLG	R†	G	Yes	Melting
RMLS	R†	S	Yes	Melting
SACC	S,R†	C	Yes	Accretion
SACI	S	I	Yes	Accretion
SACR	G,S	R	Yes	Accretion
SAUI	S	I	Yes	Autoconversion
SDPV	S	V	0§	Deposition
SDTV	S	V	0§	Deposition on ice that grows into snow
SRDI	S	I	Yes	Riming/deposition changes ice into snow
SRIC	S	C	Yes	Riming of ice crystals
VEVC	V	C	No	Evaporation
VEVR	V	R	No	Evaporation
VSUG	V	G	0§	Sublimation
VSUI	V	I	No	Sublimation
VSUS	V	S	No	Sublimation
CPHA‡	C	A	No	Phoretic attachment
CBRA‡	C	A	No	Brownian attachment
IPHA‡	I	A	No	Phoretic attachment
SICA‡	S	A	No	Inertial capture

*C=Cloud water, G=Graupel, I=Cloud ice, R=Rain, S=Snow, V=Vapor, A=Interstitial aerosol
†Temperature > 0°C
‡Scavenging only
§There is no scavenging associated with this process.

perform the saturation adjustment with respect to liquid, even at temperatures below 0°C, and ice crystal growth by deposition where we use the growth rates at water saturation from Koenig (1971) rather than assuming that deposition is a part of the saturation adjustment process. For further information see Molenkamp and Bradley (1990).

4. PARTICLE SCAVENGING PARAMETERIZATION

The scavenging parameterization is a parallel representation to the cloud microphysics with six classes of aerosol corresponding to the five types of hydrometeors and vapor. Particles ingested into a cloud can be incorporated in hydrometeors by condensation or deposition nucleation or attachment. Once they are collected we assume they are well-mixed within and move with the hydrometeors. Uncollected or unattached particles are called interstitial aerosol. The collected particles are eventually either deposited on the ground with falling hydrometeors or resuspended when the hydrometeors evaporate or sublimate.

For aerosol particles associated with each type of hydrometeor there is a prognostic conservation equation similar to (1),

$$\frac{\partial \chi_j}{\partial t} = - V \cdot \nabla \chi_j - \frac{1}{\rho}\frac{\partial}{\partial z}(U_j \chi_j \rho) + \nabla \cdot K_m \nabla \chi_j + Y_j, \tag{2}$$

where χ_j is the mass mixing ratio of aerosol associated with hydrometeor type j, and Y_j is the net rate of production of aerosol of type j. The transfer processes included are shown in Figure 3 and listed in Table 1.

4.1 Proportional Scavenging

For many of the aerosol transfer processes the rate is proportional to the water transfer rate because of the well-mixed assumption and is given by

$$Y_{jppk} = \frac{P_{jppk}}{q_k} \chi_k . \tag{3}$$

These processes are indicated in the proportional scavenging column of Table 1 and by solid lines in Figure 3.

4.2 Condensation Nucleation Scavenging

For soluble and wettable particles the most efficient mechanism for incorporating aerosol into hydrometeors is for the particles to serve as condensation nuclei. We assume that particles can serve as condensation nuclei only when they pass through a cloud boundary with an upward component of velocity since that is the time they are most likely to experience their highest supersaturation. Within the cloud additional condensation is assumed to occur on the already existing droplets, so no additional particles serve as nuclei. The preferred method for estimating the fraction of newly ingested aerosol incorporated into droplets upon entering the cloud is based on the CAMP detailed microphysical model (Edwards and Penner, 1988; Edwards, 1989; Chuang *et al.*, 1990, Penner *et al.*, 1990), where calculations over a range of updraft speeds and aerosol concentrations and solubilities appropriate to the situation being simulated are used to construct a table of the fraction nucleated. When such a table is not available, we estimate the fraction nucleated from the peak supersaturation, s_{max}, for a natural continental aerosol (Twomey, 1959)

$$s_{\max} = 0.0025\ w^{5/8}, \tag{4}$$

where w is the updraft in m/s. Using this estimate of the peak supersaturation in the activation zone, the fraction of ingested aerosol transferred to cloud droplets, f_N, is estimated by (Molenkamp, 1977)

$$f_N = 1 - (1 + C_N s_{\max})\ \exp(-C_N s_{\max}). \tag{5}$$

where f_N increases linearly for small values of $C_N s_{max}$ and approaches 1 exponentially as $C_N s_{max}$ becomes large. Values of the parameter C_N near 1000 give f_N near 1 and values less than 100 activate only a small fraction of the aerosol.

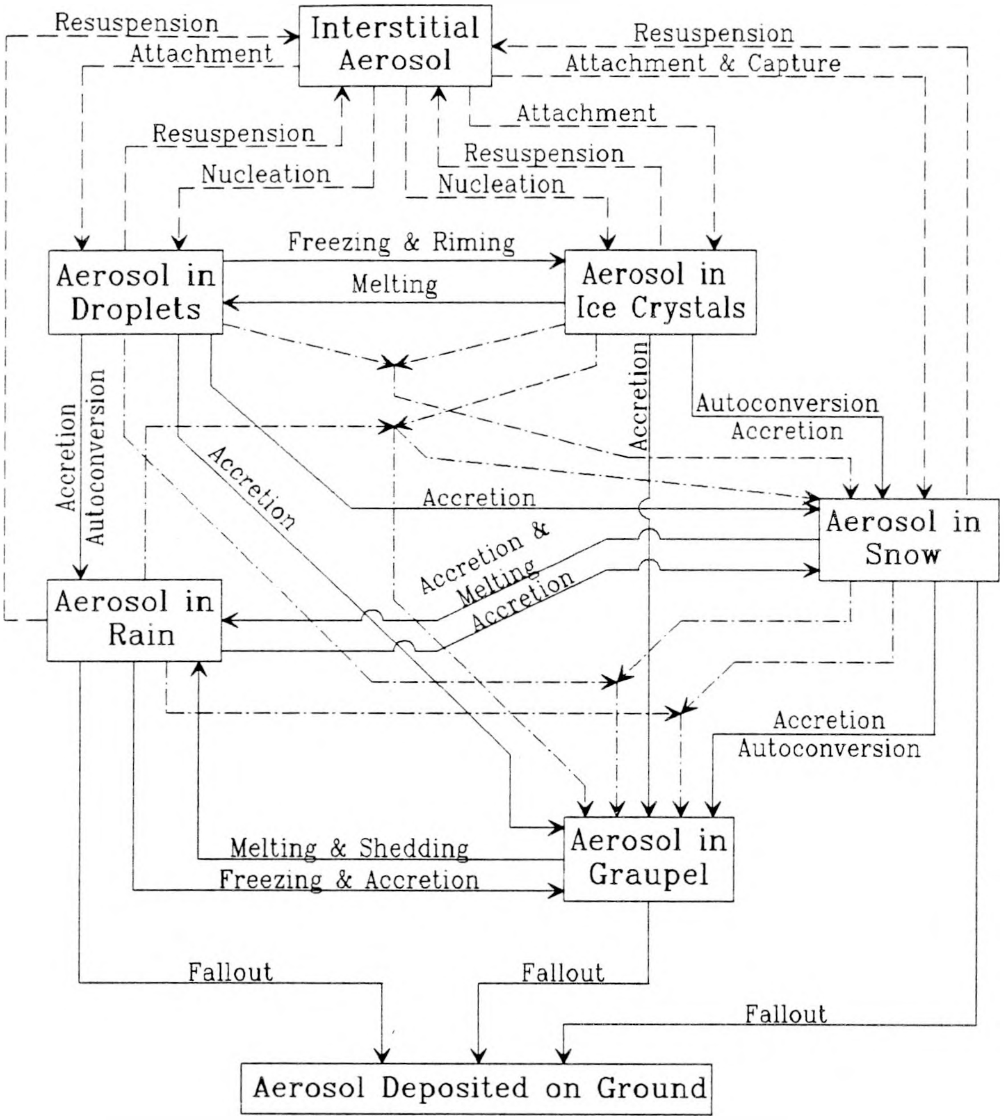

FIGURE 3. Scavenging transfer processes included in the parameterization. Solid lines indicate processes with transfer rates proportional to the cloud microphysical transfer rates.

4.3 Deposition

The deposition of water vapor on ice crystals, snow and graupel is equivalent to the growth of cloud droplets by condensation. Assuming that all the deposition occurs on hydrometeors already present, there is no scavenging of aerosol particles associated with deposition. Currently scavenging of aerosol particles that serve as deposition nuclei is not included.

4.4 Wet Graupel Growth and Shedding

Falling graupel can accrete cloud and rain water more rapidly than the collected water can freeze, especially at temperatures just below 0°C. In this case the portion of the collected liquid that does not freeze is shed as rain. We assume that the fraction of accreted pollutant that is shed with rain is equal to the fraction of collected water shed.

4.5 Resuspension by Evaporation

For a rain drop or cloud droplet that evaporates completely, the collected aerosol is resuspended as an interstitial particle, but for a drop that only partially evaporates the collected aerosol remains with the drop. Since much of the water evaporated comes from partially evaporating drops, resuspension occurs more slowly than evaporation.

Cloud droplets. In the model evaporation of cloud droplets occurs for two reasons, 1) droplets are transported outside the cloud boundary and 2) ice crystals form and grow by deposition inside the cloud tending to reduce the vapor pressure. By keeping track of the cloud water mixing ratio before and after the saturation adjustment step, the fraction of water that evaporates in a time step, f_{EV}, is known. But f_{EV} can also be written as the sum of the mass of water lost from droplets that evaporate completely plus the mass lost from larger partially evaporating droplets,

$$f_{EV} = \frac{\int_0^{r_s} m(r)\,n(r)\,dr + \int_{r_s}^{\infty} \Delta m(r)\,n(r)\,dr}{\rho q_c}, \tag{6}$$

where r is the droplet radius, $n(r)$ the droplet size distribution, $m(r)$ the mass of a droplet of radius r, $\Delta m(r)$ the mass lost by an evaporating drop of radius r during the time step, Δt, q_c the cloud droplet mixing ratio, and r_s the radius of the largest droplet that can evaporate completely in a time step. $\Delta m(r)$ can be determined by integrating the diffusional growth equation over one time step (Molenkamp and Bradley, 1990). The fraction of the aerosol in droplets that is resuspended is equal to the fraction of cloud water in droplets that evaporate completely, f_{EC}, which is given by the first term in (6).

If we assume the cloud droplets have a Khrgian-Mazin size distribution (Pruppacher and Klett, 1978), f_{EC} can be written as (see Molenkamp and Bradley, 1990)

$$f_{EC} = 1 - \exp(-\gamma_c r_s) \sum_{k=0}^{5} \frac{(\gamma_c r_s)^k}{k!}, \tag{7}$$

where γ_c is a parameter of the size distribution which can be related to q_c, and f_{EV} is

$$f_{EV} = 1 - \frac{\gamma_c^6}{120} \int_{r_s}^{\infty} \left[r^2 - r_s^2\right]^{3/2} r^2 e^{-\gamma_c r} dr. \tag{8}$$

Unfortunately (8) cannot be integrated analytically to provide an equation for r_s, but it can be integrated numerically for a set of values and used with (7) to produce a table that relates the fraction of aerosol resuspended to the fraction of water evaporated. This relationship is shown in Figure 4.

Rain. Using a technique similar to Molenkamp (1977), the rate of resuspension of aerosol due to evaporation of rain, Y_{VEVR}, is

$$Y_{VEVR} = \frac{\chi_r}{\rho q_r \Delta t} \int_0^{D_e} m(D)\, n(D)\, dD, \tag{9}$$

where $m(D)$ is the mass of a drop of diameter D, $n(D)$ the Marshall-Palmer drop size distribution and D_e the diameter of the largest drop that evaporates completely during the time step. D_e, estimated from the diffusional growth equation, is

$$D_e = \left[\frac{8(1-S)\Delta t}{\rho_w \left(\frac{L_v^2}{k_a R_v T^2} + \frac{1}{\rho q_{sw} \psi} \right)} \right]^{1/2}, \tag{10}$$

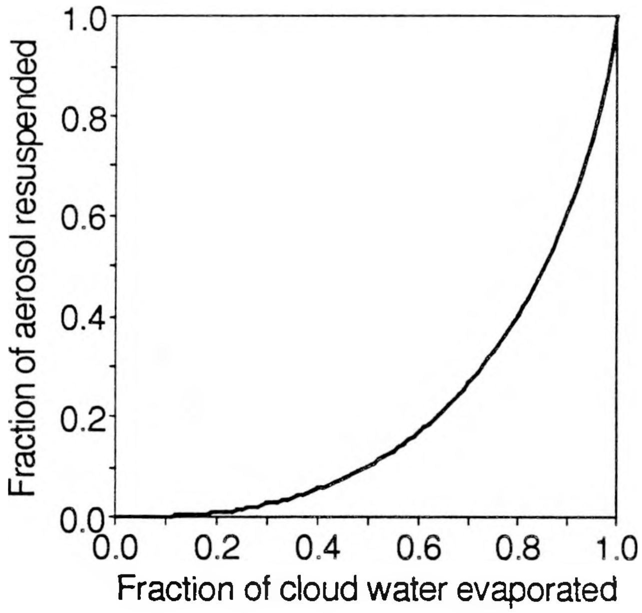

FIGURE 4. Fraction of aerosol in cloud water resuspended as a function of cloud water evaporated.

where S is the saturation ratio (less than 1 for evaporation), ρ_w the density of liquid water, L_v the latent heat of vaporization, k_a the thermal conductivity of air, R_v the specific gas constant for vapor, T the temperature, q_{sw} the saturation mixing ratio with respect to liquid, and ψ the vapor diffusivity in air. Integration of (9) over the Marshall-Palmer size distribution gives

$$Y_{VEVR} = \frac{\chi_r}{\Delta t}\left\{1 - \exp(-\lambda_r D_e)\left[\frac{\lambda_r^3 D_e^3}{6} + \frac{\lambda_r^2 D_e^2}{2} + \lambda_r D_e + 1\right]\right\}, \tag{11}$$

where λ_r is the slope parameter in size distribution. The fraction of aerosol in rain resuspended because of evaporation is very small unless the rain mixing ratio is also small.

4.6 Resuspension by Sublimation

When frozen hydrometeors move into regions where the vapor pressure is below ice saturation sublimation occurs, and aerosol is resuspended from hydrometeors that sublimate completely. Resuspension by sublimation of graupel is ignored since very few graupel particles sublimate completely before they reach the ground.

Ice crystals. The parameterization of aerosol resuspension due to sublimation of ice crystals is similar to resuspension due to evaporation of cloud droplets and rain, with the resuspension rate, Y_{ASUI}, given by

$$Y_{ASUI} = f_{ir}/\Delta t, \tag{12}$$

where f_{ir} is the fraction of the aerosol in ice crystals that sublimate completely. While all cloud droplets are assumed to have the same mass fraction of scavenged aerosol, it seems likely that small ice crystals will have larger aerosol mass fractions than larger crystals because the larger crystals probably formed earlier on nuclei active at warmer temperatures, but not necessarily of larger size, and have been growing by deposition longer without collecting more aerosol. To take this effect into account we assume a $1/r_i$ dependence for the mass fraction of aerosol in ice crystals. Then f_{ir} is

$$f_{ir} = \int_0^{r_{is}} n_i(r_i)\frac{C}{r_i}m(r_i)\,dr_i \Bigg/ \int_0^{\infty} n_i(r_i)\frac{C}{r_i}m(r_i)\,dr_i, \tag{13}$$

where $n_i(r_i)$ is the size distribution of ice crystals of mass mean radius r_i, $m(r_i)$ is the mass of the assumed spherical ice crystal, C a constant of proportionality and r_{is} the mass mean radius of the largest ice crystal that completely sublimates in a time step. The radius is again determined from integration of the diffusional growth equation,

$$r_{is} = \left[\frac{2(1-S_i)\Delta t}{\rho_i\left(\dfrac{L_s^2}{k_a R_w T^2} + \dfrac{1}{\rho q_{is}\psi}\right)}\right]^{1/2}, \tag{14}$$

where S_i is the saturation ratio with respect to ice saturation, ρ_i the density of ice crystals, L_s the latent heat of sublimation and q_{is} the saturation mixing ratio with respect to ice.

We could find no useful information on the size distribution of ice crystals, so we use a Khrgian-Mazin distribution,

$$n_i(r_i) = A_n r_i^2 \exp(-\gamma_i r_i), \tag{15}$$

where the constants A_n and γ_i can be related to the total number and mass mean radius of ice crystals. Integration of (13) under these assumptions gives

$$f_{ir} = 1 - \exp(-\gamma_i r_s)\left[1 + \gamma_i r_s + \frac{(\gamma_i r_s)^2}{2} + \frac{(\gamma_i r_s)^3}{3!} + \frac{(\gamma_i r_s)^4}{4!}\right]. \tag{16}$$

Snow. In the model snowflakes are considered to be ice crystals that have grown to sizes with mass mean diameters greater than 100 µm and aggregates of ice crystals. Since none of these snowflakes would sublimate completely in a time step, there would be no resuspension of aerosol due to snow sublimation, but Oraltay and Hallett (1990) have observed that small pieces often break off sublimating ice crystals. These small pieces are likely to sublimate completely, resuspending aerosol. Although we do not represent this breakup/resuspension process specifically, we have allowed for resuspension from sublimating snow by assuming that aerosol in snowflakes with final diameters smaller than 100 µm is resuspended. Since the mass mean diameter of snowflakes is considerably larger than 100 µm except when the snow mixing ratio, q_s, is very small, this leads to small fractions resuspended. Using the diffusional growth equation for falling snowflakes, the initial diameter of a snowflake whose final diameter is 100 µm, D_s, is given by

$$D_s^2 = (100\mu\mathrm{m})^2 + \frac{8(1-S_i)}{\rho_s(A_i + B_i)} f_v(110\mu\mathrm{m})\Delta t, \tag{17}$$

where ρ_s is the density of snow and $f_v(110\mu\mathrm{m})$ the ventilation factor for a snowflake with mass mean diameter of 110 µm. The ventilation factor is actually a function of snowflake size, but the change over the relevant size range is small and assumed constant to simplify the integration.

The fraction of aerosol in snow resuspended because of sublimation, f_{sr}, is determined by integration of the snow mass over the size distribution from diameter 0 to D_s,

$$f_{sr} = 1 - \exp(-\lambda_s D_s)\left[1 + \lambda_s D_s + \frac{(\lambda_s D_s)^2}{2} + \frac{(\lambda_s D_s)^3}{6}\right], \tag{18}$$

where λ_s is the slope parameter for the snow size distribution.

4.7 Brownian Capture

Direct capture of aerosol particles by hydrometeors can occur by Brownian, phoretic, inertial, and electrical processes. So far we have ignored electrical effects in the model although they could produce significant collection in some cases. The collection rate of interstitial aerosol by hydrometeor *j* via process *pp*, Y_{jppa}, is given by

$$Y_{jppa} = \int_0^\infty \Lambda_{pp}(a)\, m(a)\, n(a)\, da, \tag{19}$$

where a is the aerosol radius, $m(a)$ the mass of a particle of radius a, $n(a)$ the size distribution of the aerosol, and $\Lambda_{pp}(a)$ the scavenging coefficient, which is given by

$$\Lambda_{pp}(a) = \int_0^\infty K_{pp}(r,a)\, n_j(r)\, dr. \tag{20}$$

Here r is the radius of the hydrometeor, $n_j(r)$ the hydrometeor size distribution and $K_{pp}(r,a)$ the collection kernel for capture mechanism pp.

The collection kernel for Brownian diffusion is (Pruppacher and Klett, 1978)

$$K_{BR}(r,a) = 4\pi r D_p(a) f_v(r,a), \tag{21}$$

where $f_v(r,a)$ is the mean ventilation coefficient and $D_p(a)$ the diffusivity of the particles. Calculation of Λ_{BR} for the five different types of hydrometeors and evaluation for aerosol particles as small as 0.01 µm under typical atmospheric conditions reveals that only cloud droplets collect a non-negligable amount of aerosol. The scavenging coefficient for Brownian capture of interstitial aerosol can be written

$$\Lambda_{BR} = \frac{2kT\left[1 + \alpha(a) N_{Kn}(a)\right]}{3\eta_a a}\left[.543\left(\frac{N_c^2 \rho q_c}{\pi \rho_w}\right)^{1/3} + \frac{.1404 N_{Sc}^{1/3}(a) g^{1/2} N_c^{1/6} q_c^{5/6} \rho^{1/3}}{\nu \pi^{5/6} \rho_w^{1/3}}\right], \tag{22}$$

where k is Boltzmann's constant, $\alpha(a)$ the Cunningham slip correction factor, $N_{Kn}(a)$ the Knudsen number of the particle, N_{Sc} the Schmidt number for the particle, η_a the dynamic viscosity of air, N_c the total number cloud droplets, g the acceleration of gravity, and ν the kinematic viscosity of air. At a temperature of 0°C, a pressure of 600 mb and a cloud water mixing ratio or 0.001, the mean lifetime of a 0.1 µm particle is 8.5 hours and of a 0.01 µm particle 11 minutes. Since Brownian capture occurs slowly relative to many other processes considered and since it is not possible to integrate the expression for Λ_{BR} over the aerosol size distribution, we assume that Brownian capture occurs at the rate calculated for 0.1 µm particles. This assumption also recognizes that there is very little mass in smaller particles and larger ones will have been preferentially removed by nucleation scavenging.

4.8 Phoretic Capture

Thermophoresis produces a net flux of particles towards an evaporating droplet while diffusiophoresis tends to repel particles. Slinn and Hales (1971) evaluated the net effect of these forces and found that thermophoresis was dominant for particles smaller than 1 µm producing net collection during evaporation. Young (1974) included this mechanism as a method of contact nucleation in the freezing of cloud droplets to form ice crystals.

Cloud droplets. The thermophoretic collection kernel for collection of an aerosol of radius a by an evaporating droplet of radius r, $K_{TH}(a,r)$, is (Pruppacher and Klett, 1978)

$$K_{TH}(a,r) = 4\pi r f_t f_h k_a (T - T_r)/P, \tag{23}$$

where f_t is the thermophoretic factor, f_h the ventilation coefficient, k_a the thermal conductivity, T_r the temperature at the drop surface, and P the pressure. The thermophoretic factor is

$$f_t = \frac{0.4\left(1 + \alpha N_{Kn}\right)\left(k_a + 2.5 k_p N_{Kn}\right)}{\left(1 + 3N_{Kn}\right)\left(k_p + 2k_a + 5k_p N_{Kn}\right)}, \tag{24}$$

where k_p is the thermal conductivity of the particle which is much larger than k_a allowing simplification of (24) to

$$f_t = \frac{\left(1 + \alpha N_{Kn}\right) N_{Kn}}{\left(1 + 3N_{Kn}\right)\left(1 + 5N_{Kn}\right)}. \tag{25}$$

The diffusiophoretic collection kernel, $K_{DF}(a,r)$, is (Pruppacher and Klett, 1978)

$$K_{DF}(a,r) = 4\pi r D_v^* f_v \left[\frac{M_w^{1/2}}{\left(x_v M_w^{1/2} + x_a M_a^{1/2}\right)}\right] \frac{M_a}{M_w \rho}\left(\rho_{v,\infty} - \rho_{s,w}\right), \tag{26}$$

where D_v^* is the modified diffusivity of water vapor in air, f_v the ventilation factor, M_a and M_w the molecular weights of air and water, x_a and x_v the mole fractions of air and water vapor, $\rho_{v,\infty}$ the vapor density of the atmosphere and $\rho_{s,w}$ the saturation vapor density with respect to liquid at the surface of the droplet. Over the range of conditions in the atmosphere the mole fraction portion of (26) can be approximated as $\varepsilon^{-1/2}$ where ε is the ratio of M_w to M_a.

Using the relationship between the heat and mass flux for an evaporating droplet,

$$4\pi r k_a f_h \left(T_r - T\right) = -4\pi r L_v D_v^* f_v \left(\rho_{v,\infty} - \rho_{s,w}\right), \tag{27}$$

the net phoretic collection kernel can be written

$$K_{DF} - K_{TH} = K_{DF}\left[1 - \frac{f_t L_v \varepsilon^{1/2}}{R_a T}\right]. \tag{28}$$

To put these rates into a form that can be used in the model it is necessary to relate the diffusiophoretic collection rate to the evaporation mass flux. From the saturation adjustment step which sets a new value for cloud water mixing ratio, $q_{c,new}$, that maintains the atmosphere at saturation when droplets are present, the mass flux of vapor to cloud droplets during condensation, W_T, (negative of evaporation) can be determined,

$$W_T = \rho P_{CCNV} = \rho\left(q_{c,new} - q_c\right) / \Delta t. \tag{29}$$

Young (1974) gives the diffusiophoretic kernel in terms of the mass flux of vapor to a droplet of radius r as

$$K_{DF}(r,a) = \frac{4\pi r^2 g_d W(r)}{m_w^{1/2}\left[n_w m_w^{1/2} + n_a m_a^{1/2}\right]}, \tag{30}$$

where m_w and m_a are the masses of water vapor and air molecules, n_w and n_a are the number concentrations of water and air molecules, and g_d is the diffusiophoretic factor which is a weak function of particle Knudsen number with an empirically determined value near 1. Integration of this collection kernel over the size distribution of cloud droplets gives for the scavenging coefficient,

$$\Lambda_{DF}(a) = \frac{1}{\rho\epsilon^{1/2}}\int_0^\infty 4\pi r^2 W(r)\,n(r)\,dr, \tag{31}$$

where the denominator in (30) has been approximated by $\rho\varepsilon^{1/2}$. Recognizing the integral in (31) as the total flux of water to cloud droplets and using (29), the net phoretic scavenging coefficient is

$$Y_{CPHA} = \left[\frac{f_t L_v}{R_a T} - \frac{1}{\epsilon^{1/2}}\right]\left(-P_{CCNV}\right)\chi_a. \tag{32}$$

Young (1974) has shown that there is a relatively small variation of the phoretic collection kernel over the aerosol size range from 0.01 to 1 µm, so we assume that all aerosols behave as 0.1 µm particles.

Ice Crystals. Using the same approach, the net phoretic attachment rate of aerosol particles to ice crystals can be written

$$Y_{IPHA} = \left[\frac{f_t L_s}{R_a T} - \frac{1}{\epsilon^{1/2}}\right] P_{VSUI}\,\chi_a, \tag{33}$$

where L_s is the latent heat of sublimation.

4.9 Inertial Capture

Calculation of the inertial capture rate of submicron aerosol particles by falling hydrometeors gave small values because of small collision efficiencies for the particles and small terminal velocities for cloud droplets and ice crystals. Inertial capture by snow yielded the highest values which were comparable to Brownian and phoretic rates; therefore, we have included inertial capture by snow and ignored it for the other hydrometeors.

Snow. The inertial capture of aerosol particles by snow is evaluated in a manner similar to the accretion of cloud water,

$$Y_{SICA} = \chi_a \int_0^\infty E(D_s,a)\,\frac{\pi D_s^2}{4}\,U_s(D_s)\,n_s(D_s)\,dD_s, \tag{34}$$

where $U_s(D_s)$ is the snow terminal velocity, $n_s(D_s)$ the snow size distribution and $E(D_s,a)$ the collision efficiency. Sauter and Wang (1989) give the collision efficiency of a 0.75 µm particle with planar type snowflakes as

$$E\left(D_s,a\right) = E_{0s}(a)\,D_s^{-e}, \tag{35}$$

where D_s is in m, and E_{0s}(0.75µm) and e are 3.28×10^{-7} and 1.28 respectively, while Knutsen *et al.* (1976) give

$$\log_{10} E(D_s,a) = 2.477 + 1.366\log_{10}\frac{a}{D_s}. \tag{36}$$

Because of the tremendous scatter in the data on which these expressions are based there is very little to suggest that one is better than the other. Since Knutsen *et al.* allows one to estimate the effect of aerosol size, we use their expression. Assuming the collected aerosols are 0.5 μm, (36) can be written in the form of (35) with $E_{0s} = 7.4\text{x}10^{-7}$ and $e = 1.366$. Performing the integral in (34) gives

$$Y_{SICA} = \frac{eE_{0s}}{4}(\pi n_{0s})^{\frac{1-d+e}{4}} \Gamma(3+d-e)\left(\frac{\rho q_s}{\rho_s}\right)^{\frac{3+d-e}{4}}\left(\frac{\rho_0}{\rho}\right)^{\frac{1}{2}}\chi_a, \tag{37}$$

where Γ is the gamma function. At a pressure of 500 mb and a temperature of -20°C the ratio Y_{SICA}/χ_a is $2\text{x}10^{-5}$ for q_s equal to 10^{-3} and $8\text{x}10^{-7}$ for q_s equal to 10^{-6}. Therefore, when the snow mixing ratio is 10^{-3}, the mean lifetime of aerosol particles for inertial capture by snowflakes is about 8.5 hours.

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DISCUSSION

C. M. BANIC. You have presented your microphysics parameterization of aerosol scavenging and included thirty-seven processes. Based on your experience with the results from your model, what are the most significant and least significant processes for aerosol scavenging?

C. R. MOLENKAMP. One of the main reasons for using a numerical model of scavenging is to differentiate between dominant, significant and negligible processes and to identify the important interactions between processes. An earlier version of this paper included information on the typical magnitudes of many of the transfer processes, especially for the attachment mechanisms, but that material was deleted because of space considerations. We are developing a capability with this model to plot the transfer rates of the various processes in three dimensions at various times during the run to understand in more detail how the precipitation and scavenging processes interact.

I find it useful to differentiate two steps in the scavenging process, the initial capture of aerosol particles and the transfer of captured pollutant to rapidly falling rain and graupel which leads to its deposition. The initial collection of soluble and wettable particles is accomplished predominantly through condensation nucleation; this process can transfer over 90% of the ingested aerosol mass into droplets. The other attachment mechanisms are only important for hydrophobic particles or when there are very large numbers of particles such as in fire plumes. The mean lifetime of aerosols before capture by these non-nucleation processes is typically several hours so that a relatively small percentage of the aerosol mass is collected during passage through a convective cloud. While the amounts may be small, if one were concerned with toxic hydrophobic particles, these attachment mechanisms could be very significant.

Once particles have been collected by cloud droplets and ice crystals they must be transferred to rain and graupel to be carried to the ground. Accretion is the most important mechanism, but other processes must produce the initial rain, snow, and graupel before accretion can occur. Most of the pollutant that is deposited on the ground is initially collected by condensation nucleation, then accreted by graupel with subsequent melting or by raindrops and carried to the ground with rain.

R. C. EASTER. Please comment on aerosol size distribution. First, what assumptions in OCTET and in the CAMP nucleation scavenging model are made regarding the aerosol size distribution? Second, what aerosol scavenging processes are most affected by using the bulk mixing ratio approach as opposed to carrying some size distribution information in the model (e.g., mean and standard deviation of an assumed log-normal distribution)?

C. R. MOLENKAMP. OCTET does not specifically include aerosol size distribution information, but many of the interactions, particularly nucleation and attachment, are dependent on aerosol size. A CAMP simulation for the Hardiman prescribed burn is described elsewhere in these proceedings (Chuang C. C., Penner J. E. and Edwards L. L., 1991, Drop size distributions and the efficiency of nucleation scavenging over the Hardiman fire, *This publication*). The CAMP model, which represents aerosol and drop size distributions using a large number of size bins, assumes a log-normal or a superposition of log-normal distributions for the ambient aerosol and measured size distribution data for emitted smoke. The parameterized expression for fraction nucleated, which is used in OCTET when CAMP data are not available, specifies the peak supersaturation based on parameters appropriate to a natural continental aerosol, but the actual fraction does not depend directly on the size distribution. The rates for the various attachment processes assume the largest particles were removed by condensation nucleation, leaving only sub-micron aerosol particles. Since the remaining pollutant mass is concentrated in the largest particles left, we assume rates appropriate to 0.1 μm particles.

There are many limitations to the bulk-water parameterization; poor representation of size distribution is one, ignoring ice crystal shape and aerosol fractionation (different pollutant concentrations in different size drops) are others. Improving representations in these areas involves adding variables and interactions to a model that is already pushing the limits of today's most powerful computers. I think we can still learn a great deal from bulk-water models, but we also need to examine microphysical interactions in more detail, perhaps in one and two-dimensional dynamic models as we have done for condensation nucleation with CAMP.

Although it is difficult to draw conclusions without comparison to results from a more detailed model, resuspension due to evaporation and vertical dispersion of drops falling with different terminal velocities seem to be the processes most affected by a lack of size distribution information. The dependence of cloud droplet number concentration on activation of condensation nuclei, which is not included in the model, is important particularly if one wants to explore the effects of aerosols on the microphysical evolution of clouds and cloud radiative effects. The number concentration of ice crystals is another parameter which is poorly represented in the model but that has a significant effect on cloud evolution and aerosol scavenging. Even with these limitations, however, the OCTET model is providing tremendous insights into the complex dynamic and microphysical interactions between aerosols and clouds.

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