Implications of droplet nucleation to mineral dust aerosol deposition and transport

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[1] Calculations from a microphysics model are shown which indicate the factors that control droplet nucleation scavenging of hydrophilic mineral dust particles over a large range of conditions including the size, chemical composition, and number density of particles in both cumulus and stratus clouds. We focus specifically on the activation threshold radius (ATR) for droplet nucleation which determines the particles that are activated and those available for further transport and subsequent iron deposition to the remote ocean. Results suggest: the ATR is typically found in the range of clay-sized particles (radius = .1 to 1. μm), a spectrum over which the amount of dust removed declines ∼60% both in surface area and particle number; nucleation of silt-sized particles (1.–10. μm) occurs under most conditions; larger fractions of mineral aerosols are removed in cumulus clouds than in stratus; and while acid coating of dust particles in polluted environments acts to decrease the ATR, the effect is reduced by competition with soluble aerosols. Regional mineral dust environments exhibit potentially diverse aerosol wet removal impacts. The ATR representative of the tropical Atlantic ocean basin (<.2 μm) indicates ∼80% removal of the total dust surface area, while in the pristine southern hemisphere mid latitudes an ATR ∼.5 μm implies ∼60%. In contrast, varying conditions in the polluted region of East Asia suggest a large ATR spectrum (.2 to 3. μm) with dust surface area removal ranging from >80% to <10%.
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1. Introduction

[2] Mineral dust particles, which are generally less than 10 μm in radius (r), are entrained into the atmosphere in desert and semi-arid regions, dispersed globally by winds and subsequently removed from the atmosphere by gravitational settling (efficient for r > 5 μm), dry deposition and wet deposition during precipitation events. Dust particles are mostly insoluble, and are effective ice nuclei in the atmosphere [*Pruppacher and Klett*, 1997; *DeMott et al.*, 2003]. This allows efficient nucleation removal during snowfall or in deep cumulus clouds for both hydrophobic and hydrophilic particles. However, it is not clear if or when hydrophilic mineral particles can be activated as droplet nuclei and what dust aerosols are hydrophilic. It has been observed that water droplets would nucleate on quartz and samples of surface dust only under high supersaturations not observed in the atmosphere [*Pruppacher and Klett*, 1997], suggesting that silt-sized particles are hydrophobic and do not nucleate cloud droplets. An earlier study indicated that clay-sized particles may be hydrophilic and removed by cloud nucleation [*McDonald*, 1964]. More recently it was found that aged dust aerosols are often coated by sulfate and nitrate through heterogeneous chemical reactions, and could become cloud condensation nuclei (CCN) similar to sulfate aerosols [*Levin et al.*, 1996]. Sulfate coating may also result from cloud processes: cloud drops collect dust particles while new sulfate is produced by gas scavenging and subsequent oxidation of SO_2 in the drops, and then evaporate [*Wurzler et al.*, 2000; *Yin et al.*, 2002]. This introduces the possibility that dust may originate as hydrophobic particles and then be transformed to hydrophilic depending on the amount of background pollution during transport.

[3] Previous modeling studies have assumed that all dust aerosols were emitted having either hydrophobic or hydrophilic properties. In contrast, *Fan et al.* [2004] hypothesized that non-reacted dust particles are hydrophobic, and that the particles may be transformed (aged) to hydrophilic by heterogeneous reactions with SO_2, H_2SO_4, HNO_3, N_2O_5 and other species. They found that Asian dust may be transformed when encountering significant air pollution in East Asia and consequently removed rapidly by wet deposition, while cleaner air over the African region allowed dust particles to remain hydrophobic longer over the tropical North Atlantic Ocean. This differential regional impact led to a marked global improvement in comparisons of model results with annual average observations of dust concentration. In addition to the possibility of dust aging, the rate of removal by droplet nucleation may be size-dependent.

[4] In light of these measurement and modeling studies, we investigate the possible size dependent removal of hydrophilic mineral dust by droplet nucleation. For hydrophilic aerosols, below-cloud scavenging is not nearly as important as nucleation removal. We present calculations of droplet nucleation and growth using a microphysics model of an ascending air parcel in cumulus and stratus clouds, considering varying chemical compositions in clean air and polluted environments. The potential ramifications of these results are then qualitatively interpreted in distinct regional areas influenced by transport and deposition of mineral dust.

2. The Microphysics Model

[5] The critical supersaturation above which condensation nuclei become activated (a condition of positive feedback between growth and air-droplet vapor gradient)

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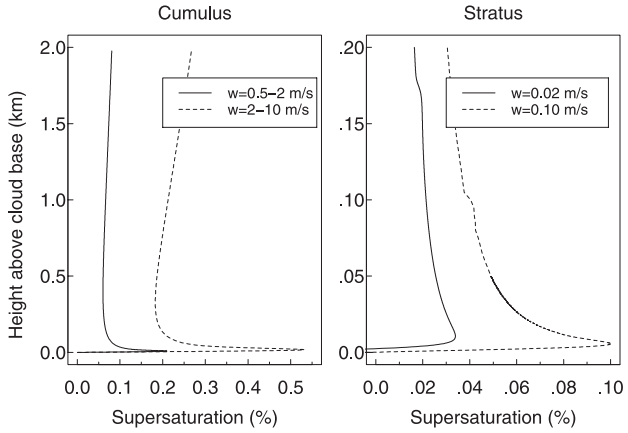

Figure 1. Time-dependent solutions for water vapor supersaturation in air parcels. The vertical velocity of each air parcel at the cloud base is indicated in the legend. Profiles are shown for model cases C05, S05, and S14 in Table 1 and for a case of rapid rising cumulus.

depends on the size and soluble mass of the particles [*Alofs et al.*, 1989]. We calculate the rate of droplet growth by water vapor condensation in clouds using a numerical microphysics model that considers both the size and chemical compositions of the nuclei in an ascending air parcel. The model is described by *Liu and Wang* [1996], which has accurate formulations of the vapor gradient, vapor and thermal diffusion coefficients. The growth of a cloud droplet is represented by the change of droplet radius r with time:

$$r\frac{dr}{dt} = \frac{1 + 0.01S - f(r)}{\frac{\rho_L R_V T}{De_s} + \frac{\rho_L L f(r)}{KT}\left(\frac{L}{R_V T} - 1\right)} \quad (1)$$

and

$$f(r) = \exp\left[\frac{2\sigma}{\rho_L R_V T r} - \frac{i\varepsilon M_W \rho_d r_d^3}{\rho_L M_S\left(r^3 - r_d^3\right)}\right] \quad (2)$$

where S is percent supersaturation, ε the mass fraction of soluble material in the nucleus, σ the surface tension of solution droplet, ρ_L density of the aqueous solution, ρ_d density of the dry nucleus, r_d radius of the dry nucleus, R_V gas constant for water vapor, T temperature of the air parcel, e_s the saturation vapor pressure, i the van't Hoff factor, M_W molecular weight of water, M_S average molecular weight of soluble materials in the nucleus, L latent energy of condensation, D diffusivity of water vapor modified for gas kinetics, and K modified thermal conductivity of moist air. The equations that describe the rates of cooling and changes in water vapor, liquid water, supersaturation and pressure in the air parcel will not be presented for brevity in this paper, but can be found in work by *Liu and Wang* [1996] and *Pruppacher and Klett* [1997]. For dust particles, the soluble fraction is assumed to be sulfate surface coating on a sphere.

[6] We include three aerosol types, sulfate, seasalt and mineral dust, each represented by 20 logarithmically equal size bins ranging from 0.01 μm to 10 μm in radius. The size distribution of sulfate is given by a single mode with a mass median radius (MMR) of 0.2 μm and a geometric standard deviation (GSD) of 2. The size distribution of dust aerosols is represented by a coarse mode (85% mass, MMR = 2 μm, GSD = 2) and a fine mode (15%, MMR = 0.4 μm, GSD = 2). The size distribution of seasalt aerosols is similarly represented by a coarse mode (80% mass) and a fine mode (20%). For the dust particles, we specify the mass fraction of soluble materials to be 0.01%, 1%, or 5% for all size bins, respectively. The observed soluble mass was about 1% at Barbados in July [*Li-Jones and Prospero*, 1998]. A relative humidity of 99% is assumed for the calculation of initial droplet sizes just below the cloud base. The equations are integrated numerically over time.

[7] There are two major precipitating cloud types; shallow and deep cumulus prevalent in the tropics and mid latitude summer, and stratus associated with ascent of air in synoptic scale weather systems. The main difference between them is the vertical velocity of the air parcel. For this study we ignore deep convection which activates all mineral dust aerosols as ice nuclei. The updraft velocity for the shallow cumulus case is specified to be 0.5 m s^{-1} at the cloud base, increasing linearly with altitude to 2 m s^{-1} at 2 km above the base [*Siebesma and Cuijpers*, 1995]. For the weaker vertical velocities of stratus clouds we chose two cases where air parcels ascend either at a constant rate of 0.02 m s^{-1} typical of common mid latitude synoptic scale low pressure systems or 0.1 m s^{-1} associated with the strongest cyclones.

3. Model Results

[8] Figure 1 shows model supersaturation (S) in the clouds as a function of height above cloud base. Maximum S values, which are reached near the cloud base, increase with the ascension velocity and are found to be about 0.035% and 0.1% for the stratus clouds and 0.21% and 0.53% for the cumulus clouds. Higher S is also maintained for more rapid rising clouds reflecting a balance between the effects of cooling, which increases S, and condensation, which decreases S. Figure 2a shows the vertical profiles of droplet radii in the shallow cumulus. Rapid droplet growth is obtained in the first 300 m above cloud base, and slower increases of radius are obtained further up. A cloud droplet nucleated on a fine sulfate particle can grow nearly as large as a droplet nucleated on a coarse dust particle ten times its size. Similar growth curves are obtained in stratus clouds (Figure 2b), except for the larger droplet sizes afforded by lower droplet concentrations in stratus clouds.

[9] The minimum size of nucleating particles provides a transition point between dust particles available for continuing transport and those which are removed by precipitation. This may have a significant influence on the global contribution of mineral dust to the atmospheric radiative balance and the iron flux to the oceans. The ATR is determined by the maximum S obtained in the ascending air parcel, and is impacted by the soluble mass fraction of dust particles (a measure of aging). For example, Figure 3 shows the threshold sensitivity to the soluble mass fraction of dust particles in a shallow cumulus cloud. The 1:1 line in Figure 3 indicates no nucleation or growth, while activated

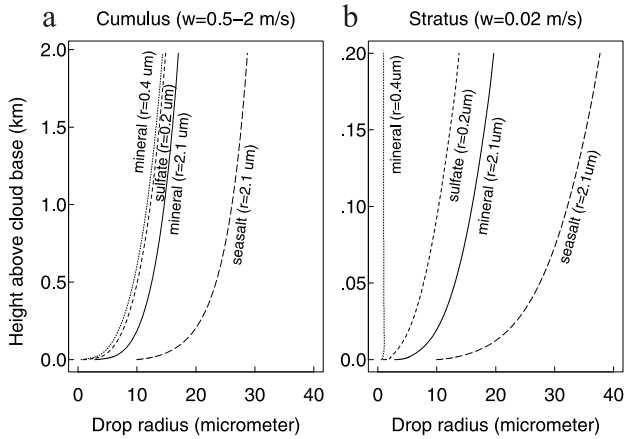

Figure 2. Time-dependent solutions for water droplet sizes (shown for nucleating particles corresponding to the mass median radius, r) in the cumulus and stratus clouds. Size profiles are shown for model cases (a) C05 and (b) S05 in Table 1.

particles grow into large droplets (r > 10 μm), as indicated by symbols located far above the 1:1 line. In this case the threshold radius is .10 μm for a sulfate coating of 1% and increases to .27 μm for a sulfate coating of .01%, indicating more clay-sized particles escape nucleation at lower soluble mass.

[10] Table 1 presents the calculated ATR over a plausible wide range of mineral dust, fine sulfate, and sulfate coating for various cloud types. Here, a mineral particle is considered activated when the droplet grows by condensation to a radius greater than twice its dry size and greater then 5 μm at 2 km above cloud base. Growth by coagulation is slow for droplets smaller than 5 μm [*Pruppacher and Klett*, 1997]. Also presented are the impact of the ATR on the percent of total surface area removed by droplet nucleation, which influences atmospheric radiation as well as downstream air-to-sea iron flux, and the percent of total particles activated, which influences available ice nuclei. Overall, while the droplet nucleation of silt-sized dust (>1. μm) occurs under most conditions, the ATR tends to vary over the scale of clay-sized particles (.1–1.0 μm). This is a range where the decrease in the amount of total dust removed is most sensitive to increasing ATR, both in terms of surface area (∼95% to ∼35%) and particle number (60% to 1%). In addition, larger fractions of mineral dust are removed in shallow cumulus than in the stratus clouds, because a greater supersaturation is reached in the former (Figure 1). Comparing cases (S05, S08), (S06, S09) or (C05, C08) indicates that the ATR increases with the number of competing sulfate aerosols. But it is insensitive to the concentration of mineral dust aerosols, as shown by cases (C04, C10), (C05, C11) and (S05, S11). Also, in polluted environments, enhancing the sulfate coating results in an increase of nucleating particles and a concomitant increase in surface area removal (e.g., cases S04–S06, C04–C06).

4. Implications

[11] The large spectrum of global environmental conditions considered in Table 1 and the resulting variability in threshold radius suggests a potentially diverse global role for droplet nucleation removal of mineral dust. By assigning appropriate parameters depicted in Table 1 to representative areas of the globe, one can postulate the regional ramifications. We broadly define three distinct regions impacted by mineral dust sources which are unique in meteorological transport, precipitating clouds, and background pollution: (1) The northern hemisphere mid latitudes, with springtime Asian dust, synoptic scale storms and stratus wet removal, and significant pollution; (2) Africa and the tropical Atlantic basin, with Saharan dust, trade wind transport, cumulus wet removal, and relatively unpolluted air; and (3) The southern hemisphere mid and high latitudes, with comparatively minor dust sources, synoptic scale transport, stratus wet removal, and unpolluted background air.

[12] Regions 2 and 3 are clean air domains. The tropical Atlantic basin (as represented by cases C01 or C02) is subjected to cumulus droplet nucleation with a threshold radius ranging from .27 μm or .10 μm, indicating that up to 40% of the total mineral dust particles could escape droplet nucleation removal and be available downwind as ice nuclei, while >80% of the total surface area would be removed. In contrast, the higher latitudes of the southern hemisphere (case S01) typically experiences stratus removal which allows dust >.5 μm to be activated. Surprisingly, while ∼60% of the dust surface area is removed, ∼97% of the total dust particles escape and can be transported downstream. This would lead to an abundance of clay-sized particles in the southern hemisphere.

[13] In region (1) the Asian dust source is located just upstream of a polluted environment. This yields significantly different transport scenarios dictated by the time in transit over SO_2 and NO sources, and the resulting amount of soluble coating (aging). Slow transport increases the soluble coating (rapid aging) which decreases the threshold radius to .38 μm (case S06), while conversely, rapid transport (less coating/slow aging, case S04) greatly increases the ATR to the silt-sized range (3.0 μm). Interestingly, this substantial increase in ATR has little impact on the number of activated

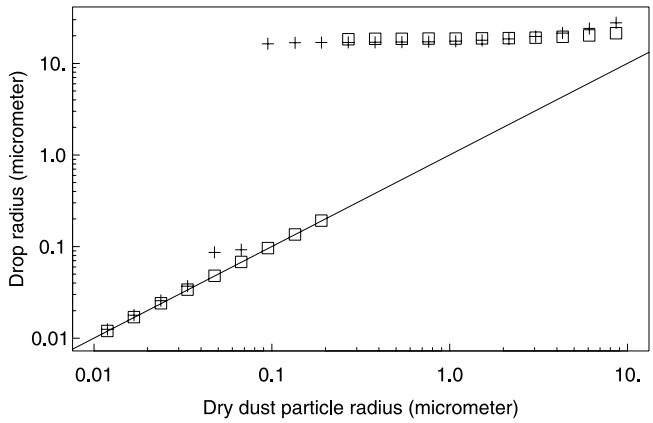

Figure 3. Snapshot of droplet sizes at 1 km above cloud base in a shallow cumulus cloud grown on nucleating mineral dust particles with soluble masses at 1% (crosses, model case C02 in Table 1) and 0.01% (squares, model case C01 in Table 1) respectively. Solid line indicates a 1:1 relationship (that is, zero growth on non-activated particles).

Table 1. A Summary of Model Inputs, Maximum Supersaturation, Activation Threshold Radii for Mineral Aerosols, Fractions of Mineral Dust Surface Area Removed, and Fractions of Mineral Dust Particles Activated by Number[a]

Model Cases	Fine Sulfate ($\mu g\ m^{-3}$)	Mineral Dust ($\mu g\ m^{-3}$)	Sulfate Coating (%mass)	S_{max} (%)	Threshold Radius (μm)	Surfaces Removed (%)	Particles Activated (%)
Stratus ($w = 0.02\ m\ s^{-1}$)							
S01	0.4	30	0.01	0.059	0.54	58	3.2
S02	0.4	30	1	0.059	0.54	58	3.2
S03	0.4	30	5	0.051	0.27	78	13
S04	4	30	0.01	0.036	3.04	7.6	0.03
S05	4	30	1	0.034	0.76	48	1.6
S06	4	30	5	0.032	0.38	68	6.5
S07	40	30	0.01	0.017	>5.0	0.0	0.0
S08	40	30	1	0.017	1.08	37	0.76
S09	40	30	5	0.017	0.54	58	3.2
S10	4	300	0.01	0.050	2.15	15	0.10
S11	4	300	1	0.032	0.76	48	1.6
S12	4	300	5	0.022	0.54	58	3.2
Stratus ($w = 0.1\ m\ s^{-1}$)							
S13	4	30	0.01	0.10	1.08	37	0.76
S14	4	30	1	0.10	0.38	68	6.5
S15	4	30	5	0.096	0.19	87	24
S16	40	30	0.01	0.044	2.15	15	0.10
S17	40	30	1	0.044	0.76	48	1.6
S18	40	30	5	0.043	0.38	68	6.5
Shallow cumulus ($w = 0.5–2\ m\ s^{-1}$)							
C01	0.4	30	0.01	0.38	0.27	78	13
C02	0.4	30	1	0.36	0.10	97	60
C03	0.4	30	5	0.34	0.07	99	77
C04	4	30	0.01	0.21	0.54	58	3.2
C05	4	30	1	0.21	0.19	87	24
C06	4	30	5	0.20	0.10	97	60
C07	40	30	0.01	0.062	1.52	26	0.31
C08	40	30	1	0.062	0.38	68	6.5
C09	40	30	5	0.062	0.27	78	13
C10	4	300	0.01	0.21	0.54	58	3.2
C11	4	300	1	0.19	0.19	87	24
C12	4	300	5	0.15	0.14	94	41

[a]Here w is the vertical velocity of the air parcel; S_{max} is the maximum supersaturation; the stratus clouds have a base temperature of 280 K at 900 mb; the cumulus clouds have a base temperature of 293 K at 930 mb; and seasalt mass concentration is 9 $\mu g\ m^{-3}$ in all cases.

particles, but a large effect on the dust surface area removal (~68% versus ~8%) which could have a profound influence on both local and oceanic iron deposition to the ocean. If there is precipitation during transport over East Asia, heavily coated particles are removed, while uncoated clay and small silt particles avoid droplet nucleation. However, if there is no precipitation all particles are transported downstream to the central and eastern Pacific. In this less polluted environment the ATR is not nearly as sensitive to soluble coating and, consequently, the surface area removal increases to >50% (cases S01–S03) enhancing wet deposition.

[14] The microphysics calculations for appropriate environmental conditions strongly suggest that the ATR is typically found within the range where droplet removal of dust, both in terms of number of particles and surface area, is highly variable. However, the global implications of droplet nucleation to mineral dust deposition and transport described here are based on qualitative assumptions. Future measurement campaigns focusing on the size distribution and hygroscopic nature of dust aerosols as well as global model transport simulations are needed to quantify the effect of variable droplet nucleation on the global distribution and deposition of mineral dust.

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