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Simple approximations for condensational growth

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Abstract
A simple geometric argument relating to the liquid water content of clouds is given. The phase
relaxation time and the nature of the quasi-steady approximation for the diffusional growth of
cloud drops are elucidated directly in terms of water vapor concentration. Spatial gradients of
vapor concentration, inherent in the notion of quasi-steady growth, are discussed and we argue
for an occasional reversal of the traditional point of view: rather than a drop growing in
response to a given supersaturation, the observed values of the supersaturation in clouds are the
result of a vapor field adjusting to droplet growth. Our perspective is illustrated by comparing
the exponential decay of condensation trails with a quasi-steady regime of cirrus clouds. The
role of aerosol loading in decreasing relaxation times and increasing the rate of growth of the
liquid water content is also discussed.

Keywords: cloud, diffusion, droplet growth

Cloud descriptions in the atmospheric science literature often begin with the observation that cloud water volume fraction f is quite small, on the order of $\times 10^{-6}$ and the distance between droplets is about hundred times their average size, (e.g., Srivastava 1989, p 870). Also, all cloud physics texts begin the topic of diffusion growth with the case of a single drop and then discuss assembly of droplets (e.g., see Rogers and Yau 1989, chapter 7). Here we shall suggest a collective view on f as a direct result of thermal interaction between drops via latent heat.

Let humid air, initially at dew point, cool suddenly by ΔT, e.g. forming a contrail. As droplets begin to form and grow by condensation, latent heat ℓ is released and raises the temperature back to T_{dew} so that no other droplet can grow in the vicinity of a given droplet. The 'vicinity' here refers to the inverse concentration, $n^{-1} \equiv V$; volume of air cell per cloud drop (inverse of number density). In other words, the system (drop) responds to environmental cooling via Le Chatelier's principle, by opposing the changes and the released latent heat is spent on warming the air 'exclusion sphere' V and the droplet itself. This yields

$$v\rho_{\text{w}}\ell = (v\rho_{\text{w}}c_{p\text{w}} + V\rho_{\text{a}}c_{p\text{a}})\Delta T \qquad (1)$$

where v is the droplet volume, ρ_{w} and ρ_{a} are water and air densities while $c_{p\text{w}}$ and $c_{p\text{a}}$ are the specific heats of water

and air densities respectively, and ℓ is the latent heat of condensation (at $0\,^\circ$C): $\ell = 2.5 \times 10^{6}$ J kg^{-1}. Neglecting heat spent on warming the droplet, and denoting $Q_{\text{in}} = m\ell$, results in

$$Q_{\text{in}} = v\rho_{\text{w}}\ell = Q_{\text{out}} \approx \rho_{\text{a}}Vc_{\text{a}}\Delta T \qquad (2)$$

where ρ and c denote density and heat capacity, respectively, and subscripts a and w, refer to air and water, respectively (dropping the subscript 'p' for pressure). Thus, for the volume fraction of liquid water in air, we obtain a convenient approximation:

$$f = v/V \approx (\rho_{\text{a}}/\rho_{\text{w}})\frac{c_{\text{a}}\Delta T}{\ell} \approx 0.4 \times 10^{-6}\Delta T \qquad (3)$$

or, in conventional units, 1 g of liquid water per kg of air for $\Delta T = 2.5\,^\circ$C. The 10^{-6} value is comprised roughly of the product of two factors of 10^{-3}. The second factor illustrates the enormity of water's latent heat of condensation ($l_{\text{vap.}}/c_{\text{air}} = 2500$ K). The volume fraction $f \sim 10^{-6}$ implies inter-drop distance about 100 average droplet diameters as is typically observed. Yet, despite this separation, the growth of the drops is collective insofar as the thermal interaction is mediated by the release of latent heat and defines the 'sphere of influence' per drop (e.g., because of lower latent heat, methane clouds can be less dilute).

So far the cooling mechanism is left unspecified (e.g., adiabatic parcel ascent, radiative cooling, or mixing). Specializing to adiabatic ascent should yield adiabatic liquid water content. In this case, temperature lapse rate $\Gamma \equiv \mathrm{d}T/\mathrm{d}z$, ΔT and the cloud depth are related via $\Delta T \approx \Gamma H$, yielding $f \approx (0.4 \times 10^{-6}\Gamma)H$. This is the volume fraction analogy of the adiabatic liquid water content and f increases with cloud height as expected. When expressed in differential form $(\mathrm{d}/\mathrm{d}z)$, this is similar to analytic expressions for the adiabatic liquid water content found in the literature (e.g., Pontikis 1996, p 2630 or Goody 1995, p 223).

We digress briefly to make two comments. The 'thermal interaction' with neighbors associates certain randomness with the notion of cloud condensation nucleus (CCN), (defined in terms of activation supersaturation only) because latent heat released by nearest neighbors may raise the temperature above the dew point (in addition to competing for water vapor), thereby depriving a given embryo of an opportunity to grow. In addition to variable vertical velocity, the thermal interaction between nearest neighbors is an alternative way to cause the 'poor empirical correlation between cloud droplet and cloud condensation nuclei concentrations', (e.g., see Chuang *et al* 1997). Indeed, the 'luck', associated in spatial proximity to one's neighbors becomes a factor in CCN potential, in addition to chemical composition and size. Also, spatial statistics of droplets can become sub-Poisson during early growth as neighbors are 'repelled' via the thermal exclusion. In calm radiative cooling conditions, this may cause increased optical extinction of fog (see Kostinski 2002, figure 3 on p 2524).

We next address the question: how quickly is this water volume fraction f attained and does the rate depend on the aerosol concentration? To that end, we begin by gaining an intuitive appreciation for the so-called phase relaxation time and for typical values of supersaturation: the reader is referred to Squires (1952) and, in particular, to Korolev and Mazin (2003), for an authoritative discussion. In contrast to prior literature, we shall work directly with concentration and will be able to avoid the $r =$ const assumption (e.g., Squires 1952, Korolev and Mazin 2003, p 2959) in deriving and interpreting the quasi-steady growth regime.

For clarity, let us examine droplets of single size and again consider a droplet of volume v, growing in a suddenly supersaturated environment, denoting $n^{-1} \equiv V$, air volume per drop. For example, at $t = 0$, T and therefore, $C_{\mathrm{eq}}(T)$, the equilibrium concentration of water vapor, is lowered as warm and humid jet engine exhaust is suddenly exposed to cold environment, and begins to form contrails. The integral mass balance gives

$$\frac{\mathrm{d}m}{\mathrm{d}t} = -\frac{1}{n}\frac{\mathrm{d}C}{\mathrm{d}t} = -V\frac{\mathrm{d}C}{\mathrm{d}t} \qquad (4)$$

where C is the average water vapor concentration in V. The drop grows in V at the expense of the diminishing concentration in V, according to mass conservation and disregarding spatial concentration gradients. On the other hand, according to the local mass conservation, the droplet grows because of the incoming water vapor flux:

$$\frac{\mathrm{d}m}{\mathrm{d}t} = 4\pi r^2\left(\frac{D}{r}\right)(C - C_{\mathrm{eq}}) = 4\pi r D \Delta C \qquad (5)$$

where C_{eq} is again the equilibrium concentration, maintained at the growing droplet surface (at $t = 0$, $C_{\mathrm{eq}} < C$). The Maxwellian expression $(D/r)\Delta C$ for the vapor flux j for a sphere was used (no time lag for diffusion) and we neglected Laplace and Raoult's terms and the effects of latent heat (see below). The condition for validity of the Maxwellian flux (quasi-stationary approximation) is $1/r(\mathrm{d}r/\mathrm{d}t)\tau_{\mathrm{diff}} \ll 1$, that is, the droplet boundary moves 'slowly' w.r.t. to the diffusion time $\tau_{\mathrm{diff}} \propto r^2/D$. The sufficient condition is $\rho_{\mathrm{vapor}} \ll \rho_{\mathrm{water}}$ (Twomey 1977, pp 71–74) and is practically always satisfied in cloud physics. More importantly, note that it is the *difference* $(C - C_{\mathrm{eq}})$ rather than the *ratio* (supersaturation) $(C - C_{\mathrm{eq}})/C_{\mathrm{eq}}$ that drives the diffusional growth.

As C_{eq} is constant in time in this case (it was suddenly changed at $t = 0$ but remains constant thereafter because ambient temperature and pressure are assumed independent of time), $\mathrm{d}C/\mathrm{d}t = \mathrm{d}(C - C_{\mathrm{eq}})/\mathrm{d}t$. Then, equating the right-hand-sides of equations (4) and (5) leads to simple exponential relaxation of quantity $\phi \equiv (C(t) - C_{\mathrm{eq}})$ to zero (saturation), with the characteristic time constant

$$\tau = \frac{1}{4\pi r D n}. \qquad (6)$$

This is the 'internal' characteristic timescale of the system (assembly of drops) response to external change.

Next, let C_{eq} change with time, in order to mimic natural variations such as adiabatic ascent or radiative cooling. This introduces an external timescale into the problem. Note that $\mathrm{d}C/\mathrm{d}t$ no longer equals $\mathrm{d}(C - C_{\mathrm{eq}})/\mathrm{d}t$, and the equation obtained by equating the RHS of equations (4) and (5) is:

$$\frac{\mathrm{d}C}{\mathrm{d}t} = -\frac{1}{\tau}(C - C_{\mathrm{eq}}) \equiv -\frac{1}{\tau}\Delta C \qquad (7)$$

where C, C_{eq}, and τ are all functions of time. Our view is that the time-dependence of C_{eq}, is prescribed by the changing environmental $P(t)$ and $T(t)$ and $C(t)$ responds to this external 'forcing' by $C_{\mathrm{eq}}(t)$ with the characteristic timescale $\tau(t)$. Then, the natural form of equation (7) is

$$\tau\frac{\mathrm{d}C(t)}{\mathrm{d}t} + C(t) = C_{\mathrm{eq}}(t) \qquad (8)$$

so that the 'driving force' is on the RHS. This is a 1st order linear ODE but with a variable and unknown coefficient as $\tau \propto [r(t)]^{-1}$ is not a known function of time. An efficient approximate scheme can be set up to solve equation (8) via the method of an integrating (exponential) factor (e.g., Boas 2006, p 401). Treating τ as a given parameter yields a 1st order solution as $C(t) = [\tau^{-1}\exp(-t/\tau)]\int C_{\mathrm{eq}}(t)\exp(t/\tau)\,\mathrm{d}t$ and the driving force $C_{\mathrm{eq}}(t)$ can often be assumed exponential (see below). Then, quasi-steady growth of $r(t)$ can render τ a variable but known function of time, etc.

Although working directly in terms of concentration has important advantages, in order to connect with the existing literature, we shall proceed in terms of the saturation ratio C/C_{eq} or supersaturation $S - 1 = (C - C_{\mathrm{eq}})/C_{\mathrm{eq}} \equiv (\Delta C)/C$. (In contrast, conventional S is defined at a point, e.g., satisfying $S = 1$ on a droplet boundary at all times.) As the 'pursuit' of $C_{\mathrm{eq}}(t)$ by C is conducted with the 'agility' τ^{-1}, one intuitively

expects asymptotic ΔC to increase with τ. To see this, divide both sides of equation (7) by C and then approximate C by C_{eq} in the denominator of the RHS as the difference is known to be less than a per cent from observations. This results in

$$\frac{C - C_{\mathrm{eq}}}{C_{\mathrm{eq}}} = -\tau \frac{\mathrm{d}[\ln(C_{\mathrm{eq}}/C_{\mathrm{ref}})]}{\mathrm{d}t} \equiv \frac{\tau}{T} \qquad (9)$$

where the logarithmic form is convenient because of the Arrhenius dependence $C_{\mathrm{eq}}(t) = C_{\mathrm{ref}} \exp{-(\ell/RT)}$. While our derivations require neither $r = \mathrm{const}$ nor $\mathrm{d}S/\mathrm{d}t = 0$, the conclusion $\Delta C/C \propto (nr)^{-1}$ is in agreement with modeling results in the literature (e.g., Rogers 1975, p 200). The quasi-steady value of $S - 1$ in equation (9) can also be understood as $\alpha\tau$, where α^{-1} is the externally imposed timescale. For the example of uniform adiabatic ascent, $\ell/RT = \ell/RT_{\mathrm{ref}}(1 - \alpha t)^{-1}$, as $T = T_{\mathrm{ref}}(1 - \alpha t)$. As $(1 - \alpha t)^{-1} \approx (1 + \alpha t)$, C_{eq} acquires exponential dependence on time, with the time constant $\alpha^{-1} = (\Gamma u)/T_{\mathrm{ref}}$, where Γ and u are the temperature lapse rate and the updraft speed, respectively. Then, $(C - C_{\mathrm{eq}})/C_{\mathrm{eq}} = (\ell/RT_{\mathrm{ref}})\alpha\tau$. The pre-factor is about $(5290 \, \mathrm{K}/T) \approx 20$. With the internal timescale τ being on the order of 20 s (see below for the effects of latent heat on τ), and α^{-1} being above 10^4 s (value for strong updrafts of several meters per second), one readily obtains a reasonable 1% order of magnitude upper bound on supersaturation.

Let us compare the above interpretation with the traditional one for quasi-steady growth (e.g., Korolev and Mazin 2003, p 2959 or Squires 1952), that is the condition of steady supersaturation, $\mathrm{d}S/\mathrm{d}t = 0$, typically interpreted as a relative humidity increase balanced by the depletion of water vapor via droplet growth. The condition $\mathrm{d}S/\mathrm{d}t = 0$ states that the ratio of $C(t)$ to $C_{\mathrm{eq}}(t)$ becomes independent of time. Working in reverse, it follows from our equation (7) that this regime is satisfied when $C_{\mathrm{eq}}(t)$ depends on time exponentially (if the weak dependence of τ on t is neglected). As we have just seen, this is the correct external driving for uniform adiabatic ascent where $C_{\mathrm{eq}}(t)$ acquires exponential dependence on time. From this perspective, conditions for quasi-steady growth are not so constraining. Also, this is in agreement with the picture of the 'pursuit' of $C_{\mathrm{eq}}(t)$ by C with the 'agility' τ^{-1}, because in the case of exponential dependence the system can lag behind and yet stay in constant ratio to the forcing. Indeed, if C lags behind $C_{\mathrm{eq}}(t)$ by τ, then, for large t, $C(t) \propto \exp(-\alpha(t - \tau))$ while $C_{\mathrm{eq}}(t) \propto \exp(-\alpha t)$ and the ratio is $1 + \alpha\tau$ for small $\alpha\tau$. Hence, we once again obtain: $S - 1 \propto \alpha\tau$.

How does the release of latent heat by the growing droplet affect the quasi-steady growth regime? By warming the surrounding air, it softens the rate of adiabatic cooling (e.g. for dry adiabatic lapse rate to the moist one) but the exponential driving in time still holds approximately. By the Le Chatelier principle again, the emission of latent heat should oppose the external changes. Indeed, it does, by increasing the relaxation time τ as the latter is no longer entirely system-dependent but acquires external parameters as follows:

$$\tau = \frac{1 + (\gamma \ell D)/k}{4\pi r D n} \qquad (10)$$

where $\gamma = \mathrm{d}C_{\mathrm{eq}}/\mathrm{d}T \approx 10^{-6} \, \mathrm{g \, cm^{-3} \, K^{-1}}$ in the tropospheric temperature range (a useful rule-of-thumb is doubling of C_{eq}

every 10 K), D is the diffusion coefficient and k is the thermal conductivity of air. As might be expected, the effect of latent heat is appreciable ($\gamma \ell D k^{-1} \approx 2$), roughly tripling the relaxation time. The derivation of the result (10) for τ, in addition to local mass balance (5), requires local heat balance:

$$\ell \frac{\mathrm{d}m}{\mathrm{d}t} = 4\pi r^2 \left(\frac{k}{r}\right)(T_{\mathrm{d}} - T_{\mathrm{a}}) \qquad (11)$$

where T_{d} and T_{a} are the drop and air temperatures, respectively as the drop is now warmer than the surrounding air. Then, C_{eq} in the material balance equation (5) can be evaluated as $C_{\mathrm{eq}}(T_{\mathrm{d}}) \approx C_{\mathrm{eq}}(T_{\mathrm{a}}) + (\mathrm{d}C_{\mathrm{eq}}/\mathrm{d}T)(T_{\mathrm{d}} - T_{\mathrm{a}})$. To get from temperature to concentration difference in equation (11) and get the modified τ from equation (5), introduce $\beta = (C_{\mathrm{a}} - C_{\mathrm{eq}}(T_{\mathrm{d}}))/(C_{\mathrm{a}} - C_{\mathrm{eq}}(T_{\mathrm{a}}))$ so that $\mathrm{d}m/\mathrm{d}t = 4\pi r D \beta (C_{\mathrm{a}} - C_{\mathrm{eq}}(T_{\mathrm{a}}))$. Then, noting that $\ell = k(T_{\mathrm{d}} - T_{\mathrm{a}})/D(C_{\mathrm{a}} - C_{\mathrm{eq}}(T_{\mathrm{d}}))$ so that $T_{\mathrm{d}} - T_{\mathrm{a}} = \ell(D/k)(C_{\mathrm{a}} - C_{\mathrm{eq}}(T_{\mathrm{a}}))$ leads to the modified expression (10) for τ. Equation (10) decouples basic thermodynamic properties of the 2nd term from those of the system (size and concentration). Environmental properties such as an updraft and lapse rate, enter α in the expression for quasi-steady supersaturation.

Having included the effects of latent heat, we return to physical interpretation of the quasi-steady growth regime. As discussed above, uniform adiabatic ascent yields $C_{\mathrm{eq}}(t) \propto \exp(-\alpha t)$, thereby providing 'driving conditions' appropriate for quasi-steady growth so that the 'system response', $C(t)$ remains in constant ratio to C_{eq}, the former lagging behind the latter. Perhaps, the most important sense in which the growth is quasi-steady is the steadiness of the spatial pattern and gradients of concentration. The physical picture is that of growing drops with $C(\mathbf{x}, t) = C_{\mathrm{eq}}(\mathbf{x}, t)$ maintained at the drop boundary at all times and $C(\mathbf{x}, t)$ far away (many drop diameters), exceeding the boundary value by $\Delta C \propto \tau$ as given by equation (9).

We now turn to interpreting these results in terms of supersaturation. This transition is not quite as trivial as might appear at first sight as the notion of supersaturation $S - 1 = \Delta C/C_{\mathrm{eq}}$ combines the 'driving force' $C_{\mathrm{eq}}(t)$ and the 'system response' $C(t)$. Insofar as it is the observed values of supersaturation $S - 1 = \Delta C/C_{\mathrm{eq}}$ that are *maintained* by the growing drops, phrases such as 'a drop growing in response to a supersaturated environment' are imprecise because supersaturation away from the droplet is already implied by the growth. In addition, S cannot be regarded a local quantity within the quasi-steady growth approximation because $S - 1 = 0$ at the drop boundary at all times but several droplet radii away, $S - 1 = \Delta C/C$ as given by equation (9). Thus, the spatial variation is prescribed rather then independently varying at each spatial point as might appear from its definition as the ratio of pressures or concentrations. Also note the associated gradient of temperature: a growing drop is ≈ 0.058 K warmer at supersaturation of 0.3%, i.e., practically never exceeding a tenth of a degree in still air.

In view of the above remarks, the frequent interpretation of a droplet, growing in a given supersaturation field (e.g., Twomey 1977, p 97) can be viewed as follows: there is a

spatial gradient of S, given by equation (9) and maintained by the quasi-steady growth of droplets. Therefore, the notion of 'microscopic supersaturation' (e.g., Srivastava 1989, p 869), that is, 'supersaturation in the vicinity of a drop rather than a macroscopic supersaturation' is misleading because the spatial gradient required by the very process of growth, 'occupies' at least several drop radii. To 'smooth out gradients of supersaturation between drops . . .' would contradict the notion of growing drops as well as the thermal 'exclusion sphere' as discussed above. In this sense, the conventional separation in cloud physics texts into individual growth followed by collective growth discussion (e.g., see Rogers and Yau 1989) can be modified to emphasize that the quasi-steady value of S is determined by the collective growth.

Note also, that the notion of supersaturation is fundamentally of limited spatial resolution, quite aside from the quasi-steady growth regime. Consider, for example, the statement $S = C/C_{eq} = 0.1\% = 10^{-3}$; a numerical value often used in articles and texts (e.g., Rogers and Yau 1989, p 88) or the discussion of kinetic limitations in Chuang *et al* (1997). Then recall that the relative fluctuation of the number of molecules n (actual rather than average value of C) in a given volume scales as $n^{-1/2}$ (Poisson law), requiring, in turn, a cubic micron of air (10^7 molecules) or so, to attain the required accuracy in S. Hence, 1 μm spatial fuzziness of S is implied, rendering the notion of microscopic supersaturation questionable at such scales.

In order to illustrate the possible importance of the perspective suggested here, consider the so-called supersaturation puzzle (Gao *et al* 2004, Peter *et al* 2006). This refers to unexpectedly high and unexplained relative humidities (RH) of 130% up to 300% observed in cirrus clouds and jet contrails. Gao *et al* (2004) give the range of 5–40 min for the contrail age at the time of observation.

Note that from the present perspective, emphasizing initial conditions directly in terms of concentration, the contrail case is essentially different from the cirrus cloud case. Indeed, the former corresponds to the exponential decay of ΔC with T and, therefore, $C_{eq}(T)$ suddenly and drastically drops (once ejected from the aircraft engine) while the latter involves gradual decrease of C_{eq} with time. Furthermore, in the contrail case, taking environmental temperature of 202 K (see Gao *et al* 2004) and initial jet exhaust temperature of about 350 K, yields the initial $S \sim 10^4$.

The question then is: how long does it take for the enormous initial supersaturation of the hot jet exhaust air to decay down to, say, 1.3 (RH of 130%)? Taking a droplet concentration of 10^2 cm^{-3} and $r = 1$ μm, (Gao *et al* 2004, p 517), we obtain $\tau \approx 2.5$ min for the relaxation time. Within the range of 5–40 min for the contrail age, this yields 2.5 to 13 (1/e) times. Note that $10^4 \times e^{-10} \approx 3.2$ so that, for example, a 25 min old contrail may still leave one with RH of 300%.

In contrast, in cirrus clouds, the initial value of S is not nearly as high but the equilibrium value $S \propto \tau \sim n^{-1}$ is larger as the number concentration is much lower. In fact, according to equation (9), the quasi-steady value of $(S - 1) \sim \alpha \tau \sim \Gamma u/nr$ is in the 10–100% range for low concentrations of a few ice particles per liter. Furthermore, such concentrations

yield relaxation times from hours to days so that the observed cirrus clouds may still be relaxing.

We now turn to examining the role of aerosol concentration in drop growth by condensation, e.g., when comparing marine and continental clouds or the effect of pollution on urban clouds. To that end, observe that the relaxation time τ depends on concentration via equation (10). As $\tau \propto (nr)^{-1}$, response of the system is more nimble as the relaxation time decreases with increasing concentration. On the other hand, according to equation (9), the observed quasi-steady supersaturation $S - 1 \propto \tau$, decreases with increasing concentration. This is in agreement with results in Korolev and Mazin (2003, equation (14), pp 2959–2960) if r is identified with the integral radius.

To isolate the effect of concentration more clearly, let the initial droplet radius be r_0 and compare two clouds with the same initial liquid water content or volume fraction f but with different number concentrations. Then, for the rate of increase of cloud water $n(dm/dt) \propto nr_0$ while keeping nr_0^3 constant, we obtain

$$n\frac{dm}{dt} \propto r_0 n \propto n^{2/3} \tag{12}$$

so that the finely dispersed cloud begins to grow faster. Similarly, the relaxation time $\tau \propto (r_0 n)^{-1} \propto n^{-2/3}$ when nr_0^3 is held constant. Thus, the finely dispersed system has a greater specific surface area and is more nimble (shorter τ). Hence, the lag between $C_{eq}(t)$ and $C(t) \sim \exp(-\alpha(t - \tau))$ is shorter, resulting therefore, in a lower quasi-steady supersaturation, despite the faster growth.

What is the nature of condensational growth before the quasi-steady regime is reached? Here aerosols play a critical role as well. The phase relaxation time $\tau[r(t)]$ depends on the radius of the growing droplet and is on the order of 10 s for concentrations of 100 cm^{-3} and $r = 10$ μm but on the order of minutes during the early growth when droplets are smaller: $r \approx 1$ μm. Thus, during the first few seconds of an 'early growth' period, one can regard $C_{eq}(t)$ decaying exponentially in time because of, say, air cooling due to uniform adiabatic ascent and C not reacting yet, as vapor field depletion due to droplet growth can be neglected. In this regime, $C - C_{eq}$ increases rapidly as $1 - e^{-t} \sim t$. As a crude estimate, this regime exists until $\tau(r(t)) \approx t$ is reached by the growing droplets.

Neglecting water vapor depletion due to droplet growth may seem to contradict the frequently made statement that droplet growth is rapid for small droplets and slows down later. Observe, however, that the implied measure of droplet growth is dr/dt, while the more suitable rapidity criterion for our problem is the rate of mass growth $dm/dt \propto d(r^3)/dt$. Also note that the ubiquitous parabolic growth $r \sim t^{-1/2}$ is based on constant S and does not hold for the early growth regime. Instead, $r^{-1}(dm/dt) \propto t$, as shown in the previous paragraph, so that $r \sim t$ during the early growth regime.

The $r \sim t$ scaling may have implications for the vexing puzzle of size-distribution broadening. Indeed, droplet surface area no longer grows linearly with time. Unlike the classic parabolic regime (e.g., Srivastava 1991, p 1597), the difference of square radii of two growing drops (variance) does not

remain constant during growth (as the mean of size distribution increases), and therefore, the relative width of the size distribution need not narrow. Furthermore, the thermal exclusion, as discussed above, causes non-uniformity in the timing of growth initiation. Finally, if the size distribution is sampled over long enough distance so that updraft fluctuations become significant, the $r(t) \propto t$ dependence renders the subsequent growth more sensitive to such fluctuations.

To conclude, we suggested a geometric (thermal exclusion sphere) perspective on the liquid water content in clouds. We then examined the phase relaxation time and quasi-steady growth in terms of simple albeit possibly crude approximations. One can see, for example, that quasi-steady growth is an exact solution for the case of exponential 'driving' of C_{eq} in time: a case of rather general applicability. Our simple expression for the quasi-steady supersaturation, $S-1 = \frac{\tau}{T}$, based on equation (9), decouples environmental properties (updraft velocity and lapse rate) entering T, from cloud properties (size and concentration), entering the denominator of τ and from thermodynamic values, entering the 2nd term in τ. We also suggested a resolution for the 'supersaturation puzzle' by treating contrails and cirrus clouds as dissimilar: exponential relaxation versus exponential driving, respectively. Finally, we illustrated the new perspective by viewing a finely dispersed 'polluted' cloud as a more nimble one (shorter relaxation time τ) so that the lag between $C_{eq}(t)$ and $C(t) \sim \exp(-\alpha(t-\tau))$ is shorter, resulting therefore, in a lower quasi-steady supersaturation for such a cloud (compared with cloud with the same liquid water content but with fewer larger drops), despite the faster acquisition of liquid water in such a cloud.

Acknowledgments

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