

SEMI-ANALYTICAL REFINEMENT OF SUBMICRON DROPLET GROWTH BY CONDENSATION

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Abstract: Understanding the growth dynamics of water droplets is crucial for accurate modelling of cloud formation and climate processes. This paper delves into the theoretical aspects of condensational growth of tiny water droplets in humid environments, such as warm clouds. The effect of droplet size on growth is examined using a semi-analytical model based on established kinetic principles, including the effects of diffusion and the medium discontinuity. While it was previously understood that smaller sizes are followed by slower growth rates, the refined model predicts that submicron droplets should grow even more slowly than anticipated. The model is consistent with previous conclusions and encompasses the growth of larger droplets as a limiting case. This model is expected to be applicable across a broad range of settings, from near-freezing conditions in clouds to elevated temperatures in technical applications involving hot steam-droplet mixtures, where Stefan flows are significant.

Keywords: condensational growth, droplet, kinetic theory, Knudsen layer, precipitation formation, cloud physics.

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1. Introduction

The primary process responsible for the formation of atmospheric clouds is the condensation of water vapour. The total mass of water vapours in the atmosphere is roughly 10^{16} kg, which is equivalent to ten thousand cubic kilometres of bulk water, with an average annual variation of this value of around 10% [Trenberth *et al.*, 2005]. Despite it comprises only 1% of the atmosphere's mass, clouds and fogs constantly cover about two-thirds of the Earth's surface, significantly affecting its radiation exchange with outer space.

The process of condensation has garnered considerable interest from researchers for both fundamental exploration and practical applications such as the development of systems for extracting moisture from the atmospheric air and artificial influencing weather patterns [Kasparian *et al.*, 2012; Kozlov, 2013; Wilderer *et al.*, 2017]. Today, numerous studies in cloud microphysics address various issues, though, a few approaches have been employed to study condensation. Direct computer modelling techniques have gained prominence in recent years [Wang *et al.*, 2019; Wang *et al.*, 2022; Wang *et al.*, 2024], but thermodynamic [Gabyshev, 2025; Quan *et al.*, 2014; Wang *et al.*, 2023a,b] and kinetic approaches [Gabyshev *et al.*, 2020; Guerrini *et al.*, 1990; Seaver, 1984] remain fundamental. This study applies the kinetic approach, which is based on classic molecular and mechanical concepts of the structure of gases and condensed matter [Eucken, 1930; Fuchs, 1959; Shuleikin, 1968].

The present study focuses on the initial droplet sizes ranging from several tens of nanometres (close to the molecular diffusion regime) to single tens of micrometres (when

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near-surface layer effects become negligible and growth rate asymptotes to the radius-square-law [Jakubczyk *et al.*, 2012; Kolwas *et al.*, 2019]). Thereby, our paper aims to provide better insights into the very initial precipitation stages in clouds before gravitational coagulation becomes prominent (prevailing at diameters $\gtrsim 30 \mu\text{m}$).

The main objective is to advance the analytical theory of stationary condensational growth of small droplets at scales comparable to the mean free path in air, assuming the absence of heat release by the condensed phase. Our semi-analytical approach elucidates the formal relationship between smaller droplet sizes and slower growth rates. Although this behaviour is generally understood, our calculation articulates and implements a specific concept underpinning it, which, despite being intuitively grasped, has not been expressed explicitly until now. While the literature may have implicitly acknowledged this concept, explicit discussion may have been avoided due to its perceived self-evidence. Nonetheless, a thorough review of the literature did not uncover any solution presented in precisely the same form as ours.

Exploration within the transition range of scales between kinetic molecular diffusion and diffusion in the developed Knudsen layer regime will show us that the growth rate of the droplets on Aitken nuclei (approximately $0.1 \mu\text{m}$ and smaller) not only deviates significantly from the radius-squared law limit but is also considerably slower than previously thought. Aitken nuclei fall within the so-called Greenfield gap [Poydenot *et al.*, 2024], a size range where particles are inefficiently removed from the atmosphere through scavenging by raindrops. This results in these particles remaining suspended for extended periods, ranging from weeks to months. The reduced growth rate of droplets on such nuclei, coupled with Brownian motion of theirs, may therefore contribute to their prolonged residence time in the atmosphere.

2. Basic assumptions and equations

Let a spherical droplet of radius r be in an infinite air space of constant temperature and humidity. The ordinary diffusion coefficient D of water vapour from the ambient air towards the droplet surface is affected by kinetic effects within the Knudsen layer, which surrounds the droplet and has a thickness approximately equal to the mean free path $\langle\lambda\rangle$ [Fuchs, 1959; Jakubczyk *et al.*, 2012]:

$$D_{\text{eff}} = \frac{D}{\frac{r}{r+\beta\langle\lambda\rangle} + \frac{D}{r\langle v\rangle\alpha_c}},$$

where β is a dimensionless coefficient that measures the thickness of the Knudsen layer in units of $\langle\lambda\rangle$ and for simplicity assumed to be taken as $\beta = 1$, noting that it is implied to follow $\langle\lambda\rangle$ in all subsequent equations; α_c is the condensation coefficient, representing the probability that the molecule transitioning to the condensed phase surface will not be reflected from it. A review of formulae for estimating the value of α_c has been provided by [Golubkov *et al.*, 2018]. The investigation of the contribution of this factor falls outside the scope of the present study. For simplicity, we therefore assume α_c to be constant, typically around 0.04 for water [Kozyrev *et al.*, 2001; Mozurkewich, 1986]. Above, $\langle v \rangle = \sqrt{\frac{kT}{2\pi m}}$ represents *one quarter* of the mean molecular speed of a water vapour molecule (with mass m). The mass flux (the increase of the droplet mass per unit time) towards the droplet surface should be determined taking into account the Stefan flow, which is responsible for hydrodynamic entrainment of air with the vapour flow [Fuchs, 1959]:

$$I_0 = -4\pi r^2 D_{\text{eff}} \frac{c_s}{c_s - \frac{c}{M}} \frac{dc}{dr},$$

where $c_s = \frac{c}{M} + \frac{c'}{M'}$ is the total molar concentration of the medium equal to the sum of the molar concentration c/M of water vapour (with an absolute humidity c and a molar mass M) and the molar concentration of the remaining components c'/M' (as dry air with the mass

concentration c' and molar mass M'), respectively. The value of c_s differs negligibly near and far from the droplet, therefore it can be considered constant. The concentration field is assumed to be steady, i.e. only the stationary growth is considered. Readers interested in the temperature decrease associated with condensation may refer to equations (3) in [Jakubczyk *et al.*, 2012] and (6.4) in [Fuchs, 1959] for a simplified treatment. In our analysis, we neglect it and consider exclusively the isothermal case.

After transformation, integration, and substitution of the integration limits as prescribed in [Fuchs, 1959], we obtain the following expression:

$$\frac{1}{r} \ln \left[\left(1 + \frac{\langle \lambda \rangle}{r} \right)^{\frac{r}{\langle \lambda \rangle}} \right] + \frac{D}{2r^2 \langle v \rangle \alpha_c} = \frac{4\pi D \Delta c}{I_0} \quad (1)$$

with the definition introduced

$$\Delta c \stackrel{\text{def}}{=} M \left[c_{s\infty} \ln \left(1 - \frac{c_\infty}{M c_{s\infty}} \right) - c_{s0} \ln \left(1 - \frac{c_0}{M c_{s0}} \right) \right],$$

where c_0 is the absolute humidity over the droplet surface and c_∞ is that of ambient air. The following form is convenient for numerical use:

$$\Delta c \equiv c_\infty x_\infty \ln \left(1 - \frac{1}{x_\infty} \right) - c_0 x_0 \ln \left(1 - \frac{1}{x_0} \right),$$

$$x_i \stackrel{\text{def}}{=} 1 + \frac{M}{M' \chi_i}, \quad i = 0, \infty,$$

where $\chi = \frac{c}{c'}$ is the water vapour mixing ratio (ratio of the water vapour density c to that of dry air c'). The ratio χ characterizes the intensity of the Stefan flow. It is small at or below room temperature, but not negligible at high temperatures (see [Gabyshev *et al.*, 2020] for all necessary material expressions). The flux of vapour condensed on the droplet is as from the equation (1). On the other hand, the flux is as follows:

$$I_0 = 4\pi r^2 \frac{dr}{dt} \rho,$$

where ρ is the bulk water density. After transformation, separation of variables, and integration, we obtain the implicit function of the droplet radius r :

$$F(r) - F(r_0) + A(r - r_0) = \tau - \tau_0, \quad (2)$$

with the following definitions introduced

$$F(r) \stackrel{\text{def}}{=} \frac{r^2}{3} \ln \left[\left(1 + \frac{\langle \lambda \rangle}{r} \right)^{\frac{r}{\langle \lambda \rangle}} \right] + \frac{r \langle \lambda \rangle}{3} \ln \left[\left(1 + \frac{r}{\langle \lambda \rangle} \right)^{\frac{\langle \lambda \rangle}{r}} \right] + \frac{r^2}{6} - \frac{r \langle \lambda \rangle}{3},$$

$$A \stackrel{\text{def}}{=} \frac{D}{2 \langle v \rangle \alpha_c}, \quad \tau \stackrel{\text{def}}{=} \frac{t}{\frac{\rho}{D \Delta c}}. \quad (3)$$

where r_0 is the initial radius of the droplet at time t_0 . This can correspond to the radius of the condensation nucleus in heterogeneous nucleation, or the scale of inhomogeneity in homogeneous nucleation (1–20 nm). We posit that this expression is valid when the initial radius exceeds $\langle \lambda \rangle$ or is comparable to that. Otherwise, not kinetic but molecular diffusion regime takes place.

3. Micrometric droplets

If the droplet is quite large $r \gg \langle \lambda \rangle$, then the thickness of the near-surface layer can be neglected, resulting in $F(r) = r^2/2$. Substituting it to (2) yields the equation describing r as a function of time t :

$$\frac{1}{2} \left[(r + A)^2 - (r_0 + A)^2 \right] = \tau - \tau_0. \quad (4)$$

The droplet radius can be expressed explicitly from the equation (4). Thence, the surface area growth rate per unit time is the following:

$$\frac{dS}{d\tau} = 8\pi - 8\pi A \left[2(\tau - \tau_0) + (r_0 + A)^2 \right]^{-\frac{1}{2}}. \quad (5)$$

It is worth noting that hints for deriving the expression (5) can be found in the famous monograph [Fuchs, 1959]. While the author of that reasons in terms of mass increase rather than area increase per unit time, the line of reasoning is not carried through to its logical conclusion, which would have resulted in an expression (5). Nonetheless, as we see, this derivation is straightforward. A minor modification of (5) can be made, noting that (in our notation $\langle v \rangle$ is a *quarter* of the mean speed!):

$$D = \frac{4}{3} \langle \lambda \rangle \langle v \rangle.$$

Within the temperature range of 0 °C to 100 °C at atmospheric pressure, the ratio $A = \frac{2}{3} \frac{\langle \lambda \rangle}{\alpha_c}$ varies monotonously and falls between 1.9 μm and 2.9 μm based on the material expressions provided by [Gabyshev et al., 2020]. When $r \gg A$, or when the time is sufficiently large $(t - t_0 \gg \frac{A\rho}{4\alpha_c \langle v \rangle \Delta c})$, the growth rate asymptotically approaches the well-known value (below, the infinity symbol in the index indicates unlimited time and the dot above denotes differentiation with respect to ordinary time t):

$$\left. \frac{dS}{d\tau} \right|_{\tau \rightarrow \infty} = 8\pi, \text{ or } \dot{S}_\infty = \frac{8\pi D}{\rho} \Delta c. \quad (6)$$

Furthermore, in the limit where $\chi \ll M/M'$, we have $\Delta c = c_0 - c_\infty$, and the equation (6) reduces to the classical ‘Maxwellian’ form [Fuchs, 1959; Jakubczyk et al., 2012]. It is this expression is commonly referred to as the radius-square-law, as the square of the radius, representing the surface area, increases linearly with time. And the constant value (6) for large droplets leads to the common use of the derivative of the area as a measure of the rate of condensational growth.

In the temperature range where Stefan flow is weak, the expression for Δc can be expanded in a series. Then the droplet growth rate $\dot{S}$ differs from that calculated by formula (6) by the following factor [Fuchs, 1959]

$$f_s = 1 + \frac{c_0 + c_\infty}{2c_s M},$$

which at room temperature gives a correction of just over 1%. This means that it can be neglected even for warm clouds, however must be taken into account for industrial applications involving hot steam-droplet mixtures [Strizhak et al., 2017].

For a moving microdroplet, the effect of the surrounding medium can be accounted for by introducing a ventilation coefficient, which depends on relevant similarity criteria. For droplets with radii less than 60 μm in laminar flow, this coefficient can be described, for example, by the formula [Pruppacher et al., 2010]:

$$f_v = 1 + 0.108 \cdot \text{Sc}^{\frac{2}{3}} \cdot \text{Re},$$

where Re is the Reynolds number and Sc is the Schmidt number. Specifically, for a 20 μm diameter droplet moving at 0.1 m/s through saturated steam, the correction f_v is only a

fraction of a percent. Therefore, this correction can be neglected when analysing cloud conditions.

It is noteworthy that the exact laws governing droplet growth or evaporation remain of particular interest to many modern researchers, due to their practical applications at the nano- and micro-scales [Dalla Barba *et al.*, 2021; Rana *et al.*, 2019]. Therefore, we will now consider the calculation of the growth rate in the presence of significant kinetic effects, where the thickness of the Knudsen layer cannot be neglected.

4. Submicrometric droplets

Combining equations (2) and (3) and employing the apparatus of inverse derivatives, we readily obtain the following expression:

$$\frac{dr}{d\tau} = \left\{ A + r \ln \left[\left(1 + \frac{\langle \lambda \rangle}{r} \right)^{\frac{r}{\langle \lambda \rangle}} \right] \right\}^{-1}. \quad (7)$$

For large radii $r \gg A, \langle \lambda \rangle$, it yields a simplified expression:

$$\frac{dr}{d\tau} \approx \frac{1}{r}.$$

Recognising that $8\pi r \frac{dr}{d\tau} = \frac{dS}{d\tau}$, from (7) we obtain an expression for the growth rate:

$$\frac{dS}{d\tau} = \frac{8\pi r}{A + r \ln \left[\left(1 + \frac{\langle \lambda \rangle}{r} \right)^{\frac{r}{\langle \lambda \rangle}} \right]}. \quad (8)$$

When the droplet radius is large $r \gg A, \langle \lambda \rangle$, expression (8) approaches the constant value 8π . This effectively recovers the well-known classical expression (6) as a limiting case. We also could similarly derive an expression for the rate of mass increase. It mirrors expression (8), but the numerator becomes $4\pi r^2 \rho$ instead of $8\pi r$.

Together with equation (2), expressions (7) and (8) form a closed system for the precise numerical calculation of the rate of change of radius and area at any given time. When considering the inverse functions $t(r)$ and $\dot{S}(r)$, the dependence $t(\dot{S})$ is fully analytical with r serving as a connecting parameter. Therefore, this approach can be termed semi-analytical (fully analytical in one direction and numerical in the other).

5. Calculations and discussion

For the calculations, the following values were adopted at 10°C [Gabyshev *et al.*, 2020]: $\langle \lambda \rangle = 6.2 \times 10^{-8}$ m, $D = 2.3 \times 10^{-5}$ m²/s, $\langle v \rangle = 140$ m/s, $\alpha = 0.04$. Generally speaking, $A = \frac{D}{2\alpha\langle v \rangle} = 2.1 \times 10^{-6}$ m $\approx 33\langle \lambda \rangle$ represents a characteristic length scale above which a droplet can be considered quite large.

Let the initial radius be equal to $r_0 = 0.1\langle \lambda \rangle$ in a moment $\tau_0 = 0$. Since the ultimate rate (equation 6) is constant, we normalise the rates to it. Thus, we denote the ratio of the growth rate (8) to (6) for the submicron case as ξ_λ , and the ratio of the rate (5) to (6) for the micrometric case as ξ_m . Both of these functions are plotted in Figures 1, 2: ξ_λ is calculated numerically (solid line) and ξ_m analytically (dashed line). It was imperative to meticulously adhere to the proper chain of calculations. The first stage involved setting the radius for the submicron model r_λ at discrete steps, for example, $0.1\langle \lambda \rangle$ apart. Subsequently, the value of ξ_λ was computed simultaneously using (8), and the corresponding time τ_λ was determined using (2). Following this, the radius r_m in the microscopic model was calculated at the same time τ_λ using equation (4), providing insight into the size that the radius would reach within the same timeframe as the droplet from the submicron model. It was only after completing these calculations that the rate in the submicron model ξ_m was derived using the time τ_λ and the radius r_m . Only this order of calculation is strictly proper, and in Figures 1, 2, the

graphs of $\xi_\lambda(r_\lambda)$ and $\xi_m(r_m)$ are actually displayed. Note that the graphs largely coincide, except in the submicrometric range, where they exhibit a substantial divergence.

The graphs indicate that the droplet growth rate approaches the asymptotic Maxwell's value (6) rather slowly. Notably, growth rate increases significantly slower in the submicrometric size range, and this finding was not previously reported in the literature. For instance, when the droplet radius equals the mean free path $\langle\lambda\rangle$, the growth rate is approximately half that previously anticipated (Figures 1 and 2, enlarged graphs). Similarly, when the droplet radius is $10\langle\lambda\rangle$, the actual growth rate may be 3% lower than previously assumed. Specifically, a droplet forming on a condensation nucleus with a size of $0.1\langle\lambda\rangle$ (the Aitken nucleus) would require a dimensionless time of $\tau = 0.005$ according to the normal model, and $\tau = 0.006$ according to our refined model, to achieve a dimensionless growth rate of $\xi = 0.12$.

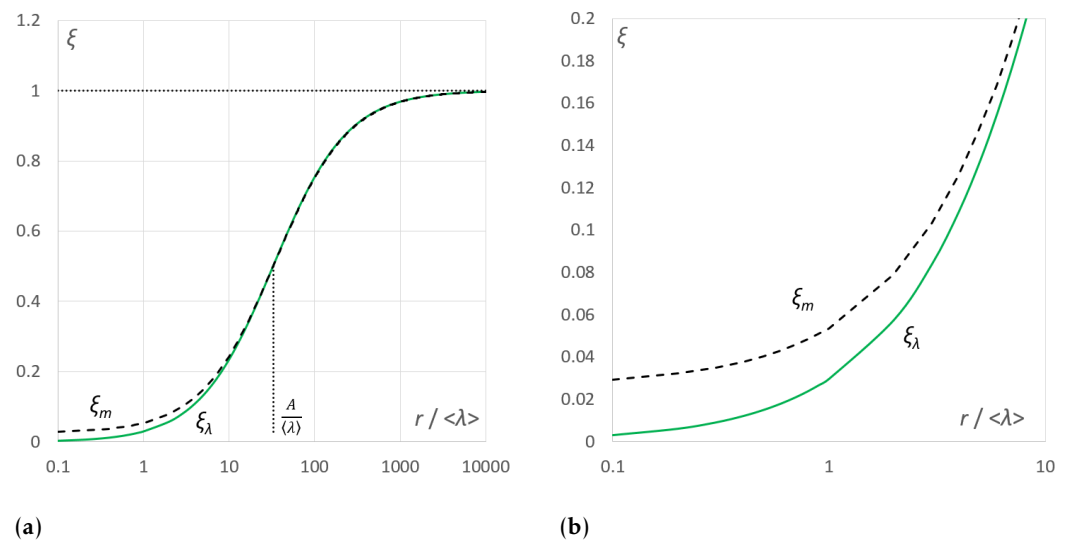


Figure 1. Droplet growth rate ξ versus radius r for the submicron model (solid line) and the microscopic model (dashed line). Enlarged graph shown right.

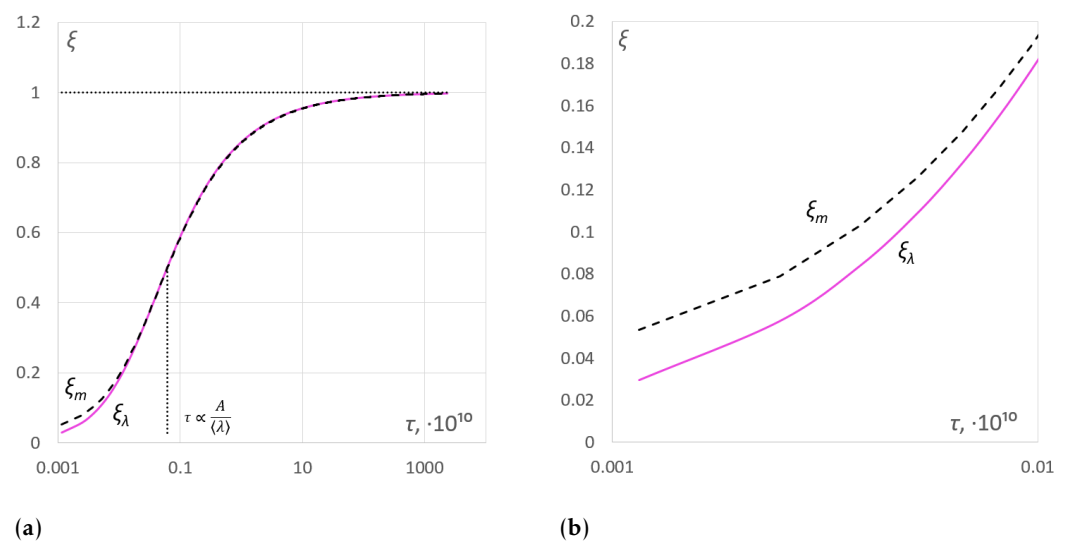


Figure 2. Droplet growth rate ξ versus time τ : submicron model (solid line), microscopic model (dashed line). Enlarged graph shown right.

6. Conclusions

Accurate assessment of droplet mass exchange is essential for understanding droplet lifetime and influencing the feasibility of airborne processes, such as atmospheric precipitation formation. This study focused on the impact of droplet size on condensation rate, motivated by the fact that cloud droplets grow by condensation to a radius of approximately 15–20 μm , beyond which coagulation prevails.

While prior research established that smaller droplets grow slower than larger ones, a definitive model that considers the transfer of molecules within the Knudsen layer and provides a clear derivation of the radius-square law, with Maxwell's growth rate as the asymptotic limit, was lacking. This investigation addressed this gap by developing a semi-analytical model based on the kinetic approach with terms that vanish at large droplet sizes. Our results demonstrate a significant impact on the growth rate, especially for submicrometric droplets. We estimated that the growth rate is roughly half of what was previously expected when the droplet radius equals the thickness of the Knudsen layer (or the mean free path, if $\beta = 1$).

It appears that other researchers might want to validate this finding using molecular dynamics simulations, and we suggest that readers consider this for future research endeavors. It is also noteworthy that our model treats concentration as a time-independent multiplier in (3) due to the steady-state approximation assumed at the outset. This enables our model to be applied even at high temperatures where Stefan flow significantly modifies the concentration factor.

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