

1 Supplementary Information:

2 1. Contributions to heterogeneous nucleation flux

3 The separation of the heterogeneous nucleation flux into “drift” and “diffusion in cluster
4 size space” contributions is described in a number of earlier works in classical nucleation theory
5 (Frenkel, 1946; Goodrich, 1964; Shizgal and Barrett, 1989; Ruckenstein and Nowakowski, 1990)
6 and textbooks (Friedlander, 2000; Seinfeld and Pandis, 2006). A similar separation is also given
7 in McGraw (McGraw, 2001). The separation of the flux following earlier works is briefly
8 described below (Goodrich, 1964; Ruckenstein and Nowakowski, 1990). The net flux from
9 clusters with g condensed molecules to those containing $g+1$ condensed molecules is (Seinfeld
10 and Pandis, 2006):

$$11 \quad J_g = \beta_g f_g - \gamma_{g+1} f_{g+1} \quad (\text{S1})$$

12 Based on the continuum approximation, J is approximated as a continuous function of g , and the
13 gradient in J_g with respect to g is given by:

$$14 \quad \frac{dJ_g}{dg} = J_g - J_{g-1} = \beta_g f_g - \gamma_{g+1} f_{g+1} - \beta_{g-1} f_{g-1} + \gamma_g f_g \quad (\text{S2})$$

15 Performing a second order Taylor expansion of the 2nd and third term of the right hand side of
16 Eq. (S2) gives:

$$17 \quad \begin{aligned} \gamma_{g+1} f_{g+1} &= \gamma_g f_g + \frac{d}{dg}(\gamma_g f_g) + \frac{1}{2} \frac{d^2}{dg^2}(\gamma_g f_g) \\ \beta_{g-1} f_{g-1} &= \beta_g f_g - \frac{d}{dg}(\beta_g f_g) + \frac{1}{2} \frac{d^2}{dg^2}(\beta_g f_g) \end{aligned} \quad (\text{S3})$$

18 Combining Eqs (S2) and (S3) gives:

$$19 \quad \begin{aligned} \frac{dJ_g}{dg} &= \frac{d}{dg}(\beta_g f_g) - \frac{1}{2} \frac{d^2}{dg^2}(\beta_g f_g) - \frac{d}{dg}(\gamma_g f_g) - \frac{1}{2} \frac{d^2}{dg^2}(\gamma_g f_g) \\ &= \frac{d}{dg}[(\beta_g - \gamma_g) f_g] - \frac{1}{2} \frac{d^2}{dg^2}[(\beta_g + \gamma_g) f_g] \end{aligned} \quad (\text{S4})$$

1 Integrating the above equation (S4) gives the desired separation of the flux into contributions
 2 from the drift in the force field and diffusion in cluster size space:

$$\begin{aligned}
 J_g &= f_g (\beta_g - \gamma_g) - \frac{1}{2} \frac{d}{dg} [(\beta_g + \gamma_g) f_g] \\
 &= \underbrace{f_g (\beta_g - \gamma_g)}_{\text{DRIFT}} - \frac{1}{2} \nabla_g \underbrace{[(\beta_g + \gamma_g) f_g]}_{\text{DIFFUSION IN CLUSTER SIZE SPACE}}
 \end{aligned}
 \tag{S5}$$

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5 **2. Comparison of GR_{drift} and GR_{cond}**

6 The size-dependent per-particle condensation rate β_g is given by (Seinfeld and Pandis, 2006):

$$\beta_g = \frac{\pi}{4} (D_p + D_m)^2 \bar{c}_\mu C_\infty
 \tag{S6}$$

8 The size-dependent per-particle evaporation rate is related to the condensation rate by (Seinfeld
 9 and Pandis, 2006):

$$\gamma_g = \frac{\beta_{g-1} C_s}{C_\infty} \exp \left[\frac{\sigma (a_g - a_{g-1})}{kT} \right]
 \tag{S7}$$

11 where a_g is the surface area of the growing cluster consisting of the initial cluster (seed) and g
 12 monomers of condensate. Letting D_s denote the size of the initial cluster, the size of the growing
 13 cluster is:

$$D_p = (D_s^3 + gD_m^3)^{1/3}$$

15 Given its small volume, addition of a monomer leads to a small increase in cluster diameter.
 16 Employing the continuum approximation at large g , we have:

$$\begin{aligned}
a_g - a_{g-1} &= \nabla_g a = \nabla_g \left[\pi (D_s^3 + gD_m^3)^{2/3} \right] \\
&= \frac{2}{3} \pi (D_s^3 + gD_m^3)^{-1/3} D_m^3 \\
&= 4 \frac{1}{6} \pi D_m^3 (D_s^3 + gD_m^3)^{-1/3} \\
&= \frac{4v}{D_p}
\end{aligned} \tag{S8}$$

2 Similarly, the gradient of the size of the growing cluster is given by:

$$\begin{aligned}
\nabla_g D_p &= \nabla_g \left[(D_s^3 + gD_m^3)^{1/3} \right] = \frac{1}{3} (D_s^3 + gD_m^3)^{-2/3} D_m^3 = \frac{\frac{\pi}{6} D_m^3}{\frac{\pi}{2} D_p^2} = \frac{v}{\frac{\pi}{2} D_p^2}
\end{aligned} \tag{S9}$$

4 Inserting equation (S8) into equation (S7), γ_g can be written as:

$$\begin{aligned}
\gamma_g &= \frac{\beta_{g-1} C_s}{C_\infty} \exp \left(\frac{\sigma \nabla_g a}{kT} \right) \approx \frac{\beta_g C_s}{C_\infty} \exp \left(\frac{\sigma \nabla_g a}{kT} \right) \\
&= \frac{\beta_g C_s}{C_\infty} \exp \left[\frac{4\sigma v}{kTD_p} \right]
\end{aligned} \tag{S10}$$

6 Combining equations (S6), (S9), and (S10), we can show that the traditional condensation
7 growth rate is essentially the contribution of drift term to the total growth rate:

$$\begin{aligned}
GR_{drift} &= (\nabla_g D_p) (\beta_g - \gamma_g) = \frac{1}{2D_p^2} (D_p + D_1)^2 \bar{c}_\mu v \left[C_\infty - C_s \exp \left(\frac{4\sigma v}{kTD_p} \right) \right] = GR_{cond}
\end{aligned} \tag{S11}$$

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1 **Supplementary References:**

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