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Supplement of

Impacts of coagulation on the appearance time method for new particle growth rate evaluation and their corrections

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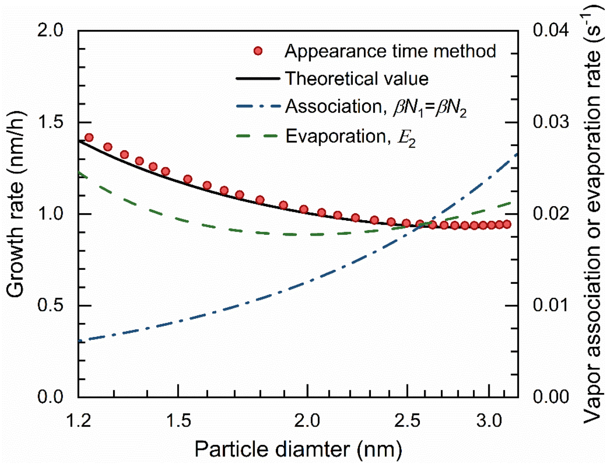

Figure S1 The growth rate estimated using the appearance time method for particle growth with a volatile vapor and a non-volatile vapor. The concentration of these two vapors, N_1 and N_2, are assumed equal and constant. β is the coagulation coefficient between the vapor and a particle and it is a function of the particle size. The evaporation rate of the volatile vapor, E_2, is assumed to be size-dependent. As indicated in the figure, the critical size for the volatile vapor is ~2.5 nm. The evolution of aerosol size distribution is simulated using a 2-dimentional discrete model. Particle coagulation is neglected in this simulation. The theoretical value of net condensation is obtained based on particle growth flux.

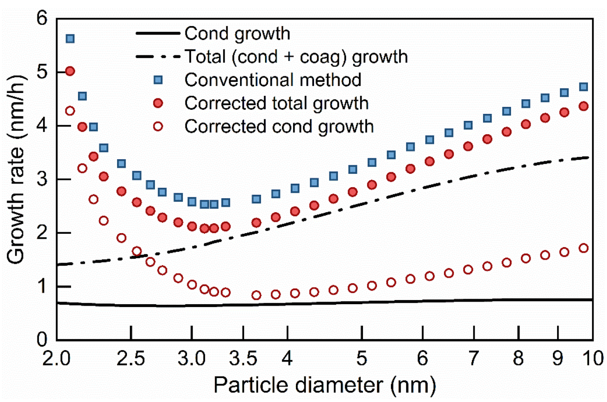

Figure S2 The appearance time method under a varying vapor concentration. The test condition is similar to that of Fig. 7, as summarized in Table A1, No. 8, and the only difference between these two tests is the size of the vapor molecule. The relative molecular masses of the vapor molecules for this test and the test in Fig. 7 are assumed to be 400 and 143, respectively. As a result, the shapes of the growth rate curves in this figure are similar to those in Fig. 7 but they are shifted towards larger particle sizes and higher growth rates.

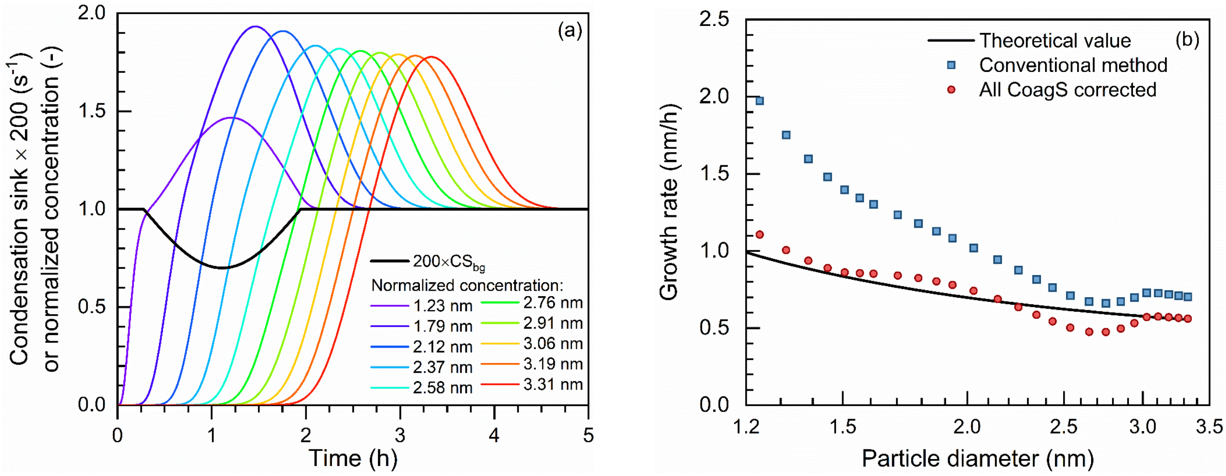

Figure S3 Influence of the variation of coagulation sink on the growth rate estimated using the corrected appearance time method. a) Condensation sink contributed by background particles and the normalized particle concentrations as a function of time. The background condensation sink (CS_{bg}) is contributed by a certain concentration of 100 nm particles, which varies with time during the growth of 1.2-3 nm particles. CS_{bg} characterizes the coagulation sink (CoagS) of particles contributed by these 100-nm background particles. The particle concentration is normalized by dividing its steady-state concentration. Due to the decrease of CS_{bg}, the maximum value of the normalized particle concentration exceeds 1.0. b) Theoretical particle growth rate and the growth rates estimated using the conventional and corrected appearance time method. The theoretical growth rate is calculated using the vapor condensation rate. The 50% appearance time is calculated using the steady-state concentrations. The coagulation sink contributed by both the 100-nm particles and new particles are accounted for when correcting the influence of coagulation sink. The deviation between theoretical growth rate and the estimated growth rate using the corrected appearance time method is mainly caused by variation of coagulation sink, which is not accounted for in the correction formula.

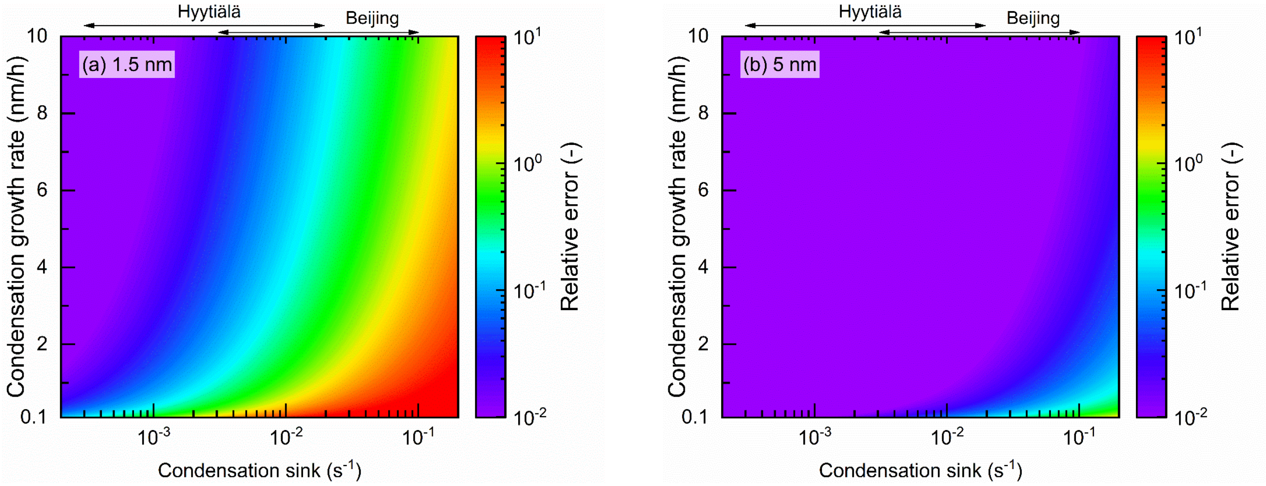

Figure S4 Error of the growth rate retrieved using the conventional appearance time method for (a) 1.5 nm and (b) 5 nm particles. The relative error is defined as $(GR_{conv}–GR_{cond})/GR_{cond}$, where GR_{conv} is the growth rate retrieved by the conventional appearance time method and GR_{cond} is the condensation growth rate. Coagulation growth is neglected in this figure. The approximate range of condensation sink in Beijing (Wang et al., 2013) and Hyytiälä (Dal Maso et al., 2002) are marked with arrows, which indicate the typical condensation sink in polluted and clean environments, respectively.

The impacts of coagulation source and their corrections

In this section, we present a derivation for Eq. 6. For the convenience of illustration, particle size and growth rate are characterized using the molecule number rather than particle diameter. Assuming that condensation is the only cause of the change in N_i (Eq. 10), the apparent growth rate is equal to the condensation growth rate, i.e.,

$$\mathrm{GR_{conv}} = \mathrm{GR_{app}}^{(10)} = \beta_{1,\mathrm{i}} N_1 \qquad \text{(Eq. S1)}$$

where $GR_{app}^{(10)}$ is the apparent growth rate (s^{-1}) of particles containing i molecules and the superscript (10) indicates the population balance assumption in Eq. 10; $\beta_{1,i}$ is the coagulation coefficient ($cm^3 \cdot s^{-1}$) between a vapor molecule and particle i; and N_i is the concentration (cm^{-3}) of particle i. N_1 is assumed to be constant. The conventional appearance time method takes GR_{app} as the growth rate (GR_{conv}) without correction. The source and maximum concentration of N_i are given below:

$$\mathrm{Src}^{(10)} = \beta_{1,\mathrm{i-1}} N_1 N_{\mathrm{i-1}} \qquad \text{(Eq. S2)}$$

$$N_{\mathrm{i},\infty}{}^{(10)} = \frac{\beta_{1,\mathrm{i-1}} N_{\mathrm{i-1},\infty}{}^{(10)}}{\beta_{1,\mathrm{i}}} = \frac{2\beta_{1,\mathrm{i-1}} N_{\mathrm{i-1}}}{\beta_{1,\mathrm{i}}} \qquad \text{(Eq. S3)}$$

where Src is the source for N_i; the $N_{i-1,\infty}$ is the maximum concentration of N_{i-1} (at $t \rightarrow +\infty$); N_{i-1} is the concentration of particle i-1 at its appearance time, hence, it is equal to 50% of $N_{i-1,\infty}$.

Now we consider the scenario with coagulation sink and coagulation source (Eq. 19). The source and maximum concentration of N_{i} become:

$$\mathrm{Src}^{(19)} = \beta_{1,\mathrm{i}-1} N_1 N_{\mathrm{i}-1} + \mathrm{CoagSrc_i} \tag{Eq. S4}$$

$$N_{\mathrm{i},\infty}{}^{(19)} = \frac{\beta_{1,\mathrm{i}-1} N_1 N_{\mathrm{i}-1,\infty}{}^{(19)} + \mathrm{CoagSrc_i}}{\beta_{1,\mathrm{i}} N_1 + \mathrm{CoagS_i}} = \frac{2\beta_{1,\mathrm{i}-1} N_1 N_{\mathrm{i}-1} + \mathrm{CoagSrc_i}}{\beta_{1,\mathrm{i}} N_1 + \mathrm{CoagS_i}} \tag{Eq. S5}$$

where $\mathrm{CoagS_i}$ is the coagulation sink ($\mathrm{s^{-1}}$) corresponding to N_{i}; and $\mathrm{CoagSrc_i}$ is the coagulation source term ($\mathrm{cm^{-3}\cdot s^{-1}}$) corresponding to N_{i}.

As illustrated in the main text, $\mathrm{CoagS_i}$ and $\mathrm{CoagSrc_i}$ change both Src and $N_{\mathrm{i},\infty}$. The appearance time and hence the retrieved growth rate are mainly influenced in two aspects: 1) the steady-state concentration and 2) the particle source that determines the time to reach a certain steady-state concentration. The conventional (apparent) growth rate under this scenario can be obtained by approximately accounting for these two aspects, i.e.,

$$\begin{aligned}
\mathrm{GR_{conv}} &\approx \beta_{1,\mathrm{i}} N_1 \cdot \frac{N_{\mathrm{i},\infty}{}^{(10)}}{N_{\mathrm{i},\infty}{}^{(19)}} \cdot \frac{\mathrm{Src}^{(19)}}{\mathrm{Src}^{(10)}} \\
&= \beta_{1,\mathrm{i}} N_1 \cdot \frac{2\beta_{1,\mathrm{i}-1} N_1 N_{\mathrm{i}-1,\mathrm{app}}}{\beta_{1,\mathrm{i}} N_1} \Big/ \frac{2\beta_{1,\mathrm{i}-1} N_1 N_{\mathrm{i}-1,\mathrm{app}} + \mathrm{CoagSrc}_i}{\beta_{1,\mathrm{i}} N_1 + \mathrm{CoagS}_i} \\
&\quad \cdot \frac{\beta_{1,\mathrm{i}-1} N_1 N_{\mathrm{i}-1,\mathrm{app}} + \mathrm{CoagSrc}_i}{\beta_{1,\mathrm{i}-1} N_1 N_{\mathrm{i}-1,\mathrm{app}}} \\
&= \left(\beta_{1,\mathrm{i}} N_1 + \mathrm{CoagS}_i\right) \cdot \left(1 + \frac{\mathrm{CoagSrc}_i}{2\beta_{1,\mathrm{i}-1} N_1 N_{\mathrm{i}-1,\mathrm{app}} + \mathrm{CoagSrc}_i}\right) \\
&= \beta_{1,\mathrm{i}} N_1 + \mathrm{CoagS}_i + \frac{\mathrm{CoagSrc}_i}{N_{\mathrm{i},\infty}}
\end{aligned} \tag{Eq. S6}$$

According to Eq. S3, the correction formula for the condensation growth rate, $\mathrm{GR_{cond}}$ is:

$$\mathrm{GR_{cond}} = \mathrm{GR_{conv}} - \mathrm{CoagS}_i - \frac{\mathrm{CoagSrc}_i}{2N_{\mathrm{i,app}}} \tag{Eq. S6}$$

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