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New particle formation contribution to aerosol mass: regimes and implications for future urban environments

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Abstract. New particle formation (NPF) is a major source of global aerosol number concentration yet its contribution to aerosol mass remains unclear. Here we propose a regime framework for NPF mass contributions using aerosol dynamic simulations constrained by atmospheric measurements. We show that new particles can contribute more than 10 μg m^{-3} to particle mass under conditions characterized by high initial new particle concentrations, high condensable vapor production rates, and low background condensation sink (CS_{bg}). Among these influencing factors, the initial CSbg governs NPF mass contribution: NPF contributes significantly to aerosol surface area and mass at an initial CSbg of ~0.001 s^{-1} (regime I), while its contribution becomes negligible when CS_{bg} exceeds 0.01 s^{-1} (regime III). Results show that NPF events in urban Beijing occurs mostly in a transition regime (initial CS_{bg} 0.001–0.01 s^{-1}, regime II), in which the NPF mass contribution is also modulated by the mass accommodation coefficient of background particles, while events at a Finnish forest site falls in regimes I and II. With a declining CS_{bg}, NPF mass contribution in urban environments will shift toward regimes I and II, indicating that control of NPF precursors represents an effective strategy for present and future particulate matter pollution.

1 Introduction

Atmospheric aerosols have profound impacts on the climate (Szopa et al., 2021) and present severe threats to human health (Lelieveld et al., 2015). Episodic haze formation is often the result of complex interactions among emissions, meteorology, and atmospheric chemistry (An et al., 2019; Huang et al., 2020; Coe, 2020). New particle formation (NPF) via nucleation and subsequent growth is well established as a major contributor to aerosol number concentration, occurring frequently in environments ranging from pristine to polluted environments (Zhang et al., 2012; Kerminen et al., 2018; Zhao et al., 2024). However, as a key formation pathway for airborne aerosols, gas-to-particle conversion via condensation and reactive uptake is commonly assumed to be dominated by large background accumulation-mode aerosols due to their large surface area. While pioneering studies have proposed associations between NPF events and haze formation(Guo et al., 2014; Kulmala et al., 2021; Kulmala et al., 2022), the roles of growing new particles in promoting gas-to-particle conversion and contributing to aerosol mass remains an open problem.

The difficulty in addressing this problem lies in both observational and conceptual dimensions. Observationally, the new particle mode after sufficient growth and background particle mode coexist at overlapping sizes (e.g., ~100 nm), making unambiguous mode separation inherently difficult. Conceptually, a prevailing hypothesis argues that gaseous precursors are converted into the particle phase independently of aerosol surface area distribution, implying that NPF is largely irrelevant to aerosol mass budgets: in the absence of NPF, the same amount of vapors would condense onto background particles. Existing theoretical frameworks based on particle survival probabilities have been successfully established to quantify NPF contributions to number concentration and CCN (Weber et al., 1997; Kerminen and Kulmala, 2002; Pierce and Adams, 2007) and later validated

https://doi.org/10.5194/egusphere-2026-4269
Preprint. Discussion started: 21 August 2026

by atmospheric measurements (Cai et al., 2022), but the progresses in similar approaches for assessing mass contributions remain limited. Kulmala et al. (2022) employed aerosol dynamic simulations with constant background condensation sink (*CS*) and fixed new particle growth rates (*GR*), showing that decreased primary emissions combined with sustained vapor emissions may enhance secondary aerosol production. However, the static treatment of vapor concentration precluded dynamic treatment of the competence of vapor uptake between growing new particles and background particles, which is fundamental in accessing NPF mass contributions.

The hypothesis that NPF is largely irrelevant to aerosol mass requires scrutiny on three aerosol kinetic grounds. First, vapor uptake is kinetically influenced process, for which an elevated surface area concentration accelerates particle mass accumulation. The surface area of new particles during subsequent growth may approach or exceed that of background aerosols, directing vapor flux toward the growing new particle mode. Second, vapor uptake by large background particles if often retarded by surface aging, organic coatings, and particle-phase diffusion (Shiraiwa et al., 2011; Renbaum-Wolff et al., 2013). These effects are characterized by effective mass accommodation coefficients (α_{bg}) substantially below unity (Kulmala and Wagner, 2001), reducing background particle competitiveness for vapor uptake relative to growing new particles. Third, observed growth rates of new particles exhibit moderate variability despite large variations in condensable vapor concentrations, particle concentrations, and *CS*, indicating an evident yet underrepresented weak response of new particle growth to the availability of measured condensable vapors (Kulmala et al., 2022; Stolzenburg et al., 2023). Additionally, for heterogeneous processes which are important for the uptake of volatile species such as N_2O_5 and SO_2 (Davidovits et al., 2006; Yang et al., 2024), the rate of vapor uptake is proportional to aerosol surface area, suggesting that NPF-derived surface area may enhance secondary mass production beyond the condensation processes limited by available condensable vapor budget.

In this study, we investigate the role of NPF in aerosol mass budgets by establishing a conceptual framework to elucidate vapor uptake competition between new particles and background particles. We also identify conditions favoring significant NPF mass contributions and the governing factors. Using a zero-dimensional aerosol dynamic model that explicitly simulates both the growing new particles and background particles, we perform parametric analyses of key influencing factors and evaluate the results against long-term observations from urban Beijing and Finnish boreal forest. The resulting regime framework provides a mechanistic basis for anticipating how NPF mass contributions will evolve and also insights for air pollution control strategies targeting secondary aerosol formation under declining primary aerosol emissions.

2 Materials and Methods

2.1 Measurements and data analysis

Particle number size distributions (PNSD) measured from urban Beijing and Finnish boreal forest were used to provide constrains on typical conditions for atmospheric NPF. Measurements in Beijing was conducted at the BUCT site (39.94°N, 116.30°E), located on the west campus of Beijing University of Chemical Technology, representing polluted environments under the influences of strong anthropogenic emissions. Long-term measurements of atmospheric pollution with a focus on particulate matter pollution and new particle formation started in 2018. PNSD ranging from 1 nm to 10 μm was monitored using a home-built diethylene glycol scanning mobility particle spectrometer system (DEG-SMPS, 1–7.5 nm) and a particle size distribution system (3 nm – 10 μm) (Liu et al., 2016; Cai et al., 2017a). Measurements at the Finnish boreal forest was conducted at the SMEAR (Station for Measuring Earth Surface – Atmosphere Relationships) II station (61.10°N, 24.17°E), located at Hyytiälä in southern Finland, representing

https://doi.org/10.5194/egusphere-2026-4269
Preprint. Discussion started: 21 August 2026

relatively clean environments. Long-term measurements at SMEAR II started in 1995 (Nieminen et al., 2014). PNSD ranging from 3 nm – 20 μm was measured using a differential mobility particle spectrometer (DMPS) (Aalto et al., 2001) and an aerodynamic aerosol spectrometer (APS, TSI 3321). Here we use the data measured until 2022 from Beijing and data in 2018–2019 from Hyytiälä. Detailed information on the measurement sites, instruments, and dataset have been reported elsewhere. (Hari and Kulmala, 2005; Cai et al., 2021; Deng et al., 2022).

At both sites, standard data quality control procedures were applied, including removal of instrument artifacts and periods with abnormal meteorological conditions. NPF events were identified using established classification criteria based on the temporal evolution of PNSD (Dal Maso et al., 2005). New particle formation rate (J) was derived using a population balance equation with improved accuracy for the influence of coagulation (Cai and Jiang, 2017). GR was derived by tracking the trajectory of the growing new particle mode using the mode-fitting method (Stolzenburg et al., 2005). The value of CS_{bg} was taken as the average total CS during the nucleation period (determined using the measured J) of every NPF event, during which the contribution of freshly nucleated particles to the total CS was negligible for their small sizes. $d_{p,end}$ of the growing mode was determined at the end of observed NPF events, characterized by a dramatical change in the mode diameter of the growing new particles due to the change in meteorology.

2.2 Model description

We employ a zero-dimensional box model to simulate the competition of vapor uptake between new particles and background particles. The model tracks two particle populations: particles originating from NPF (the new particle mode) and background particles (the background mode), together with the concentration of a single representative condensing vapor. The initial diameter of new particles is set to 5 nm, bypassing the high sensitivity of particle survival probability to the *CS* and *GR* in the sub-5 nm particle size range (Kulmala et al., 2017; Kulmala et al., 2022; Cai et al., 2022). The initial number concentration of the new particle mode represents the total number of particles produced at 5 nm during an NPF event, which is obtained by integrating the 5-nm formation rate (J_5) over the NPF period. Accordingly, the number concentration of the new particle mode can be expressed as:

$$N_{np}(d_{p,np}) = N_{np0} \cdot P(5\ \text{nm} \rightarrow d_{p,np}) \quad (1)$$

where $d_{p,np}$ is the count median diameter of the new particle mode, P is particle survival probability, and N_{np0} is the initial number concentration of the new particle mode. P is computed by calculating particle loss at every time step, accounting for both intramode coagulation (i.e., coagulation scavenging by back ground particles) and self-coagulation within the new particle mode.

The mass concentration of the new particle mode is

$$M_{np}(d_{p,np}) = m_{np} \cdot N_{np}(d_{p,np}) \quad (2)$$

where m_{np} is the mean single particle mass of the new particle mode. Since particles within the same mode are not monodisperse, we assume that these particles follow a lognormal distribution and covert $d_{p,np}$ to the diameter of mean mass using the Hatch-Choate equation (Hatch and Choate, 1929). The geometric standard deviation of the new particle mode is set to 1.7, which is obtained from observed NPF events. This value overestimates the mass of new particles at small diameters (e.g., 10 nm), but does not affect the conclusions since new particle mass contribution at such sizes is negligible. Particle density is assumed to be 1400 kg m^{-3}.(Stolzenburg et al., 2018)

Background particles are represented by a lognormal mode with an initial count median diameter of 100 nm and a geometric standard deviation of 1.7. Their initial number concentration was derived from the specified initial CS_{bg}. Self-coagulation within the background mode is included but contributes negligibly to the low particle number concentration. An effective mass

https://doi.org/10.5194/egusphere-2026-4269
Preprint. Discussion started: 21 August 2026

accommodation coefficient of background particles (α_{bg}) is introduced to account for the observed phenomenon that background particles sometimes grow more slowly than new particles, reflecting a limited vapor uptake efficiency. We also examine the sensitivity of NPF mass contribution to α_{bg}.

The budget of condensing vapors can be expressed as

$$\frac{dN_v}{dt} = Q_v - \left[CS_{bg} \cdot f(\alpha_{bg}) + CS_{np}\right] \cdot N_v \quad (3)$$

where N_v is gas-phase vapor concentration, Q_v is the vapor source (i.e., production rate), $f(\alpha_{bg})$ is the Fuchs-Sutugin correction (Fuchs and Sutugin, 1970) for vapor uptake efficiency of the background mode, and CS_{np} is the condensation sink of the new particle mode. In this study, CS_{bg} is reported at α_{bg} = 1.0, with the accommodation correction applied separately via $f(\alpha_{bg})$, such that CS_{bg} can be directly related to background particle concentration independently of uptake efficiency. A hypothetical vapor with a molar mass of 300 g mol^{-1} and a density of 1400 kg m^{-3} is used to represent the ensemble of condensable vapors.

Under simplifying assumptions, expanding particle survival probability using the Kerminen-Kulmala equation (Kerminen and Kulmala, 2002) yields an analytical expression for the relative mass contribution of new particles:

$$\frac{M_{np}}{M_{bg}} = \left(\frac{d_{p,np0} + GR \cdot t}{d_{p,bg0} + GR \cdot f(\alpha_{bg}) \cdot t}\right)^3 \cdot \frac{N_{np0}}{N_{bg0}} \cdot \exp\left[\gamma \frac{CS}{GR}\left(\frac{1}{d_{p,np0}} - \frac{1}{d_{p,np0} + GR \cdot t}\right)\right] \quad (4)$$

where M_{bg} is the background mode mass concentration, N_{bg0} is the initial number concentration of the background mode, γ is a pre-factor the Kerminen-Kulmala equation, and CS is used herein to represent the total coagulation sink of new particles. Equation 4 shows that the factors governing the relative mass contribution of NPF can be reduced to four parameters: N_{np0}, CS_{bg0} (equivalent to N_{bg0}), Q_v (determining GR), and α_{bg}. However, because the response of NPF mass contribution to these parameters can be highly non-linear, we do not peruse a closed-formed analytical solution and instead rely on numerical simulations.

3 Results

3.1 New particle growth and its impacts on aerosol surface area

Long-term observations show that NPF events routinely produce a growing particle mode, reaching sizes that significantly contribute to aerosol mass (e.g., ~100 nm) after growth for 1-2 days. At the end of analyzed NPF events in urban Beijing (see section 4.1), the growing mode reached a count median diameter ($d_{p,end}$) of 50–220 nm with an average value of 103.5 nm, and the number concentration was on average 6.8×10^3 cm^{-3} with values occasionally exceeding 10^4 cm^{-3}. The corresponding mass concentration was on average 39.7 μg m^{-3}, with values approaching ~100 μg m^{-3} in a severe haze episode (Fig. S1). At the Finnish boreal forest site (Hyytiälä site), the average number and mass concentration of the growing mode at the end of NPF event were 1.3×10^3 cm^{-3} and 5.4 μg m^{-3}, respectively (Fig. S2), consistent with the cleaner environment compared to urban Beijing (Kulmala et al., 2025). While particles in the observed growing mode also originate from primary particle emissions and transport, these observations confirm that NPF events, under favorable conditions, can produce particles with sizes that are relevant to aerosol surface area and mass. This motivates a process-level analysis in which new particles and background particles are treated explicitly as two modes (see section 4.2).

The key mechanistic question is whether NPF-derived particles can acquire significant mass from condensable vapors in competition with pre-existing background aerosols, which is fundamentally a question of the relative CS of these two competing aerosol populations. Figure 1 shows the simulated evolution of particles with a constant vapor source (Q_v) that limits the total availability of vapors. The new particle mode grows from an initial diameter of 5 nm to ~150 nm with an average GR of ~ 3 nm h^{-}

https://doi.org/10.5194/egusphere-2026-4269
Preprint. Discussion started: 21 August 2026

[1], consistent with observation results in the planetary boundary layer (Nieminen et al., 2018; Kerminen et al., 2018; Stolzenburg et al., 2023). Despite the *CS* of new particle mode (CS_{np}) being initially negligible, it increases rapidly as particles grow, surpassing the *CS* of background mode (CS_{bg}) within 6 h and becoming the dominant vapor sink in the later stage of the event. Compared to another scenario without NPF, the total *CS* with NPF increases markedly.

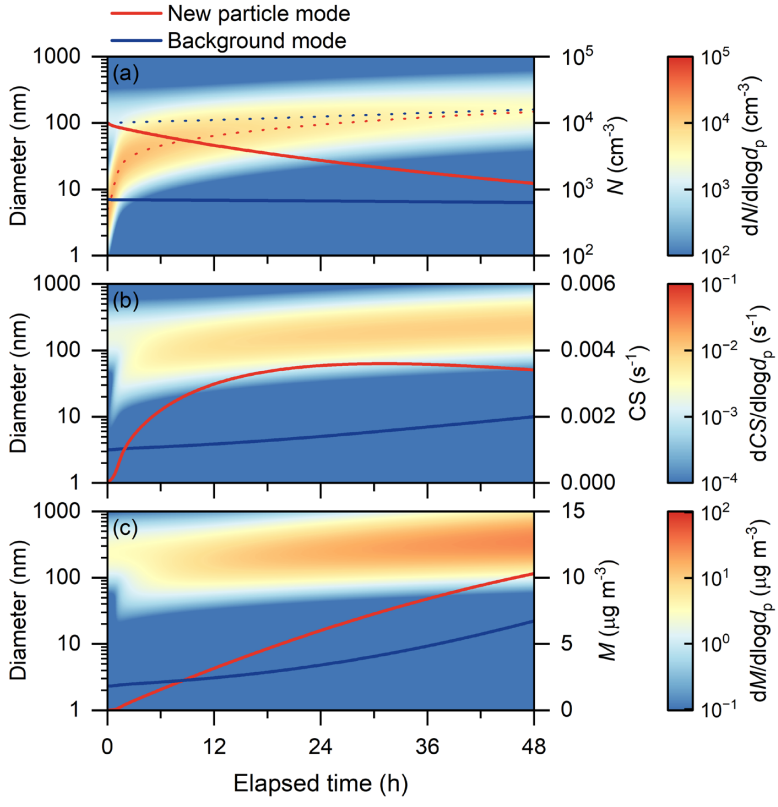

Figure 1. Simulated evolution of the new particle mode and the background mode. a) Particle number (*N*) and the number–size distribution (d*N*/dlogd_p). The dotted lines indicate the count median diameter of both modes. b) Contribution to the total condensation sink (*CS*) and the *CS*–size distribution (d*CS*/dlogd_p). c) Particle mass concentration (*M*) and the mass-size distribution (d*M*/dlogd_p). The model inputs are: constant $Q_v = 5\times10^4$ cm^{-3} s^{-1}, $N_{np0} = 10^4$ cm^{-3}, $CS_{bg0} = 0.001$ s^{-1}, and $\alpha_{bg} = 0.1$. The condensation sink of background particles in panel b is reported at $\alpha_{bg} = 1.0$ to better reflect its contribution to the surface area and the coagulation sink, which also applies to the figures below.

With a higher *CS* than the background mode, the new particle mode accumulates mass exceeding the background mode after a time lag (Fig. 1c). Similar evolution of particle size distributions, i.e., significant growth in mass-size distribution following the growth in number-size distribution, has been observed during atmospheric NPF events (Fig. S3). We note that although the constant vapor source used in Fig. 1 limited the total aerosol mass, this hypothesis holds only in the absence of heterogeneous chemistry and is not supported by atmospheric measurements. In the real world, heterogeneous chemistry can contribute additional mass preferentially on the high-CS new particle mode (Wang et al., 2010), amplifying the NPF mass contribution beyond the simulation

https://doi.org/10.5194/egusphere-2026-4269
Preprint. Discussion started: 21 August 2026

results. Figure S4 shows that there is no strong correlation between new particle growth rate and the background CS, indicating the rate of gas-to-particle conversion is not majorly limited by the competence of vapor between the two modes. Following this measurement fact, we also perform a simulation with constant vapor concentration (corresponding to a near constant growth rate), which shows a significant NPF mass contribution (Fig. S5). Nevertheless, we adopt the constant vapor source hypothesis in the following model simulations, which provide a conservative lower bound on NPF mass contributions, and focus on the relative contribution.

3.2 Regimes of NPF mass contribution

To map the conditions under which NPF mass contribution is significant or negligible, we first show the response and sensitivity of NPF mass contribution after 48 h of simulation to atmospheric conditions characterized by key influencing parameters (Fig. 2). Under the various simulated conditions, CS_{np} increases rapidly as new particles grow within ~ 12 h of simulation and drives a sustained accumulation of the mass of new particles, indicating that prolonged continuous growth is not a prerequisite for large NPF contribution to CS. Figure S6 shows that increasing the initial number concentration of new particles produced (N_{np0}) leads to a significant elevation in CS_{np} and only a moderate reduction in CS_{bg}. Interestingly, the mass of the new particle mode is insensitive to N_{np0} above ~10^4 cm^{-3} (Fig. 2b), showing that NPF mass contribution is mainly determined on the occurrence of NPF event or not but not its intensity. This is mainly because the available mass is limited by vapor production under a fixed vapor source, and also mass gains in the new particle mode are partially offset by increased scavenging of new particles by background particles, which transfers mass to the background mode.

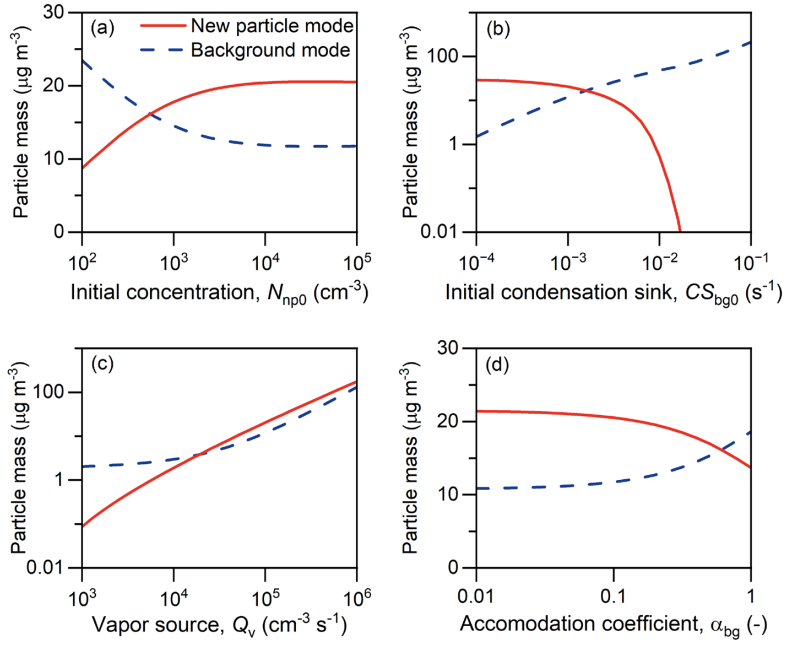

Figure 2. Influences of atmospheric parameters on NPF mass contribution after 48-h simulation. a) Influence of the initial new particle concentration produced (N_{np0}); b) Influence of the initial condensation sink (CS_{bg0}); c) Influence of the vapor source (Q_v);

https://doi.org/10.5194/egusphere-2026-4269
Preprint. Discussion started: 21 August 2026

d) Influence of the mass accommodation coefficient of background particles (α_{bg}). The model inputs are: N_{np0} = 10^5 cm^{-3}, CS_{bg0} = 0.001 s^{-1}, Q_v = 1×10^5 cm^{-3} s^{-1}, and α_{bg} = 0.1 unless a certain parameter is used as x-axis variable.

Figure 2b (and Fig. S7) shows a strong and non-linear dependence of NPF mass contribution on CS_{bg0}, which results from the non-linear response of particle survival probability to *CS* (Kulmala et al., 2017; Cai et al., 2022). Consistently, in terms of particle number that determines the occurrence of NPF events, CS has been identified as a governing factor in polluted environments such as urban Beijing (Cai et al., 2017b; Deng et al., 2021), whereas in cleaner environments such as Hyytiälä, CS has only a weak negative association with NPF occurrence (Dada et al., 2017). We could hence conclude that the critical CS threshold for significant NPF mass contribution is ~ 0.01 s^{-1} under the simulation conditions in Fig. 2b.

Sufficient vapor is a prerequisite for significant new-particle mass (Figs. 2c and S8), yet it does not have a strong influence on the relative mass contributions of the new particle mode and the background mode for Q_v above a certain threshold (~10^4 cm^{-3} s^{-1}). At the highest vapor source examined (Q_v = 10^6 cm^{-3} s^{-1}), the background mode grows rapidly and enhances the scavenging of new particles, partially offsetting the benefit of higher *GR* for new particle survival (Fig. S9).

Measurements show that the *GR* in the sub-25 nm size range tends to increase with particle size, partly attributable to the diminishing Kelvin effect (Yli-Juuti et al., 2011; Tröstl et al., 2016; Deng et al., 2020). This finding also extends to sizes over 100 nm if only monotonic growth periods are accounted for; however, the average increase in the size of background particles over the whole growth event is sometimes more slowly than smaller new particles (Paasonen et al., 2018; Dai et al., 2023). This may reflect an ineffective vapor uptake of large background particles characterized by α_{bg} below unity. Laboratory measurements have reported α_{bg} on the order of 0.01–1.0 (Davidovits et al., 2006; Liu et al., 2019). For organic-coated or highly viscous particles, the effective uptake coefficients can be substantially lower than 1.0 (Zaveri et al., 2022). Figure 2d shows that the mass contribution of the new particle mode increases as α_{bg} decreases from 1.0 to 0.1. This dependence diminishes as α_{bg} decreases further to 0.01, in which range vapor uptake is dominated by the new particle mode under the simulated condition (Fig. S10).

The findings above are summarized in Fig. 3. The results reveal a three-regime structure governed primarily by the initial CS_{bg} ($CS_{bg,0}$). One regime is characterized with significant NPF contributions. Under low background aerosol concentrations (CS_{bg0} < 0.001 s^{-1}, regime I), such as on clean days in boreal forest and in remote marine boundary layers (Zheng et al., 2021), new particles develop a mass comparable to or exceeding that of background particles. When CS_{bg0} falls below 0.0001 s^{-1}, the total aerosol mass becomes NPF-dominated with only a minor contribution from background particles. Another regime is a background-dominated regime, characterized by high background aerosol concentrations (CS_{bg0} > 0.01 s^{-1}, regime III), corresponding to polluted days in environments influenced by strong human activities. Under such condensations, new particles are rapidly removed by coagulation scavenging before they can grow to sizes with significant mass contributions. Event with the most favorable α_{bg} (0.01) representing strongly retarded vapor uptake by background particles, the contribution from the new particle mode is less than 10% (Fig. S11). In the intermediate range between these two regimes is a transitional regime (CS_{bg0} = 0.001 – 0.01 s^{-1}, regime II), in which whether the new particle mode makes a significant mass contribution is also influenced by other parameters, with α_{bg} being a key influencing factor.

https://doi.org/10.5194/egusphere-2026-4269
Preprint. Discussion started: 21 August 2026

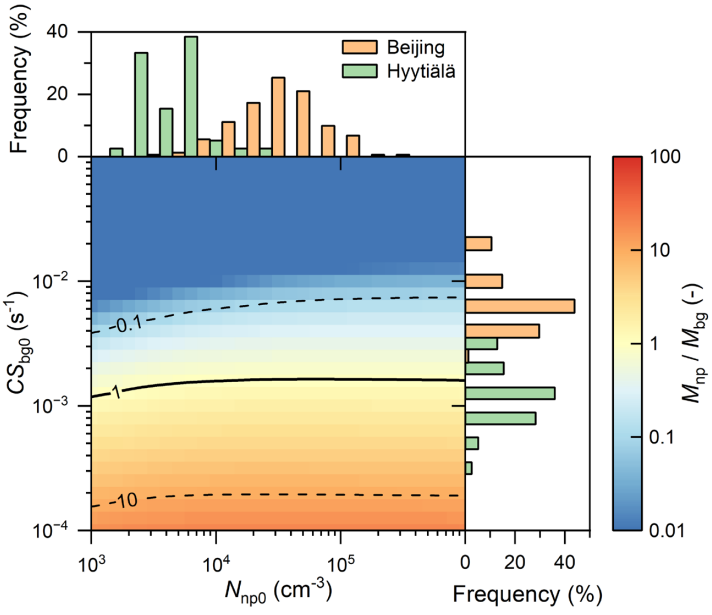

Figure 3. Contribution of the new particle mode to particle mass concentration after 48 h as a function of initial new particle concentration and background aerosol condensation sink. The other model inputs are: Q_v = 1×10^5 cm^{-3} s^{-1}, and α_{bg} = 0.1. Simulations results at different Q_v and α_{bg} values are given in Fig. S4. Statistical results of measured NPF events are given in the bar plots.

4 Conclusions and Atmospheric Implications

Positioning the observational data within the proposed regime framework quantifies the expected NPF mass contribution and its sensitivity to atmospheric variability. At both the Beijing and Hyytiälä sites, despite the difference in NPF intensities, N_{np0} often falls in the range where it exerts only a minor influence on the NPF mass contribution. That is, NPF events routinely produce sufficient seeds in the new particle mode, such that the limiting factor is the condensable vapor and particle survival probability rather than NPF intensity. The CS_{bg0} during NPF events in urban Beijing spans over 0.0026-0.019 s^{-1}, locating most events in regimes II and III. This implies the new particle mode in Beijing can occasionally, but not consistently, make a significant mass contribution. This is in accordance with both previously reported case studies on significant NPF mass contributions during haze episodes (Kulmala et al., 2021) and also the success in previous control of aerosol mass by reducing primary emissions. In contrast, CS_{bg0} at Hyytiälä falls within regimes I and II, where new particles can have a significant mass contribution after growth. This is qualitatively consistent with previous estimates that NPF accounts for a substantial fraction of cloud condensation nuclei in boreal forest environments (Tunved et al., 2006; Gordon et al., 2017; Petäjä et al., 2022).

The significant contribution of NPF to CS also has direct implications for heterogeneous processes. As discussed above, the heterogenous uptake rate scales with the available surface area. While noting that vapor uptake efficiency is also determined by the chemical compositions of the particles, which may differ between the new particle mode and background mode, Fig. S11 shows that NPF can have positive feedback on heterogeneous aerosol formation by augmenting the total particle surface area in regimes I and II. In regime III, this feedback is suppressed the dominant CS_{bg}.

https://doi.org/10.5194/egusphere-2026-4269
Preprint. Discussion started: 21 August 2026

These results carry direct implications for interpreting the importance of NPF to air quality under different atmospheric conditions (Fig. 4). In present-day polluted environments such as those in East Asia and South Asia, NPF makes a moderate and variable contribution to aerosol mass, which is governed by CS_{bg0}. This also indicates an episodic characteristic of NPF mass contributions: on relatively clean days with low CS_{bg0}, NPF events tends to occur and grow continuously during a haze episode, contributing significantly to aerosol mass but also suppress subsequent NPF events as "background particles". Similar events and feedback have been reported in previous studies (Guo et al., 2014; Westervelt et al., 2014).

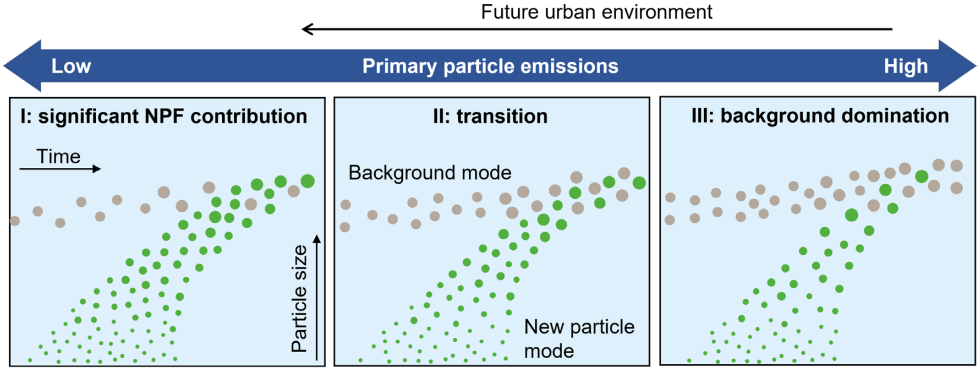

Figure 4. Schematic illustration of the regimes governing NPF mass contribution. Green and gray markers denote newly formed and background particles, respectively, with the mark size representing particle diameter. The NPF mass contribution is categorized into different regimes according to the mass fraction contributed by new particles that survive to large sizes, which is primarily controlled by the concentration of background particles. With reduced primary particle emissions in future urban environment, the relative importance of NPF mass contribution is expected to increase as a result of lower background particle concentrations.

Looking forward, sustained air pollution control strategies including industrial abatement, energy upgrade, and the transportation electrification, future urban atmospheres tend to be less influenced by primary particle emissions, shifting CS_{bg0} towards lower values. Meanwhile, due to the non-linear response of condensable vapor to unbalanced emission control (Tang et al., 2025), condensable vapor concentration may not decrease with the same extend to that in CS_{bg0}. As a result, the NPF contribution will shift from regimes II-III towards I-II, offsetting the decrease in total particle mass. This suggests that ongoing reductions in primary aerosol emissions, if not accompanied by equivalent reductions in NPF precursors, may disproportionately amplify NPF contributions to particle mass.

Data availability. Data presented in this paper are available via the link https://doi.org/10.5281/10.5281/zenodo.22037135.

Competing Interests. The authors declare no competing interests.

Author Contributions. R.C. and M.K. designed the research; R.C., S.X. and M.K. analyzed data with the help from V.-M.K.; R.C., M.K. and V.-M.K. wrote the paper with inputs from S.X.

https://doi.org/10.5194/egusphere-2026-4269
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