Responses to Reviewers' Comments on Manuscript ID ACP-2022-476

(Survival probability of atmospheric new particles: closure between theory and measurements from 1.4 to 100 nm)

We thank the editor for handling the reviews and the reviewers for the efforts and comments that help to improve this manuscript. The reviewer's comments are addressed in the following paragraphs and the manuscript has been revised accordingly. We add figures for particle growth rate in simulated cases and demonstrate that the validity of the $n_{\log}$ method is not necessarily based on a size-dependent particle growth rate. Discussions are added to address the influence of the geometric standard deviation of an aerosol population on the $n_{\log}$ method. We clarify that the closure between theory and measurements on particle survival in Beijing is mainly based on the conventional J and n methods, as they are used for sub-25 nm particles. We also added discussions on the uncertainties in the survival probability computation, including a case study of the event measured on May 20, 1998 in Hyytiälä.

The comments are shown as sans-serif blue texts and our responses are shown as serif black texts. Changes are highlighted in the revised manuscript and shown as "quoted underlined texts" in the responses. References are given at the end of the responses.

Reviewer #1

This paper presents nice closure results of the survival probability of freshly nucleated particles calculated from different approaches and its sensitivity to the measurement or derived parameters. This is an excellent piece of work and should be published in ACP.

The measured and theoretical survival probabilities are indeed sensitive to associated uncertainties and environmental variations. Authors discuss the implications on particle survival probability in measurements and models and the challenges to retrieve some of the parameters. What is the uncertainty range in particle survival probability associated with uncertainties in measurements and simulations? Can such a range of uncertainties be derived, e.g., from Figure 7?

Response: We appreciate the reviewer's comments on uncertainties and sensitivities. The discussions in section 4.3 "Uncertainties in particle survival probabilities" has been extended to better address the uncertainties in the theoretical and measured survival probabilities. Since P_{theo} is a function of exp(CS/GR), we change the definition of the sensitivity from $-\mathrm{d}P_{\mathrm{theo}}/\mathrm{d}(\mathrm{CS/GR})$ to $-\mathrm{dlog}P_{\mathrm{theo}}/\mathrm{d}(\mathrm{CS/GR})$ such that the uncertainty range of the theoretical survival probability (P_{theo}) can be readily derived using the sensitivity. We give examples on how to derive the uncertainty range of P_{theo} from Fig. 7. We have also referred to a manuscript currently being reviewed in ACPD (Tuovinen et al., 2022), which shows the uncertainty ranges of P_{theo} associated with different uncertainties in particle growth and loss rates.

The uncertainty in the measured survival probability (P_{meas}) is mainly associated with influences of emissions, transport, etc. We add a case study measured in a Finnish boreal forest to show these influences. The method to compute P_{meas} may also introduce uncertainties, yet we think these uncertainties are minor if a proper method is used according to the type of NPF events.

Revised manuscript: The sensitivity is herein defined as -dlogP_{theo}/d(CS/GR) and it can be readily computed using Eq. 3. The value of the sensitivity indicates the order of magnitude of the uncertainty in P_{theo}. For instance, for a 1.4 nm particle with CS/GR = 20 nm^{-1}, the sensitivity per nanometer growth is ~1.6, indicating that a ±10 % uncertainty in CS/GR will lead to an uncertainty factor of $10^{1.6\times10\%}$ = 1.45 (equivalent to -31% or +45% relative uncertainty) in the P_{theo} for particle growth from 1 nm to 2 nm. Similarly, the same ±10 % uncertainty in CS/GR will lead to an uncertainty factor of 3.0 (equivalent to -67% or +200% relative uncertainty) in the overall P_{theo} for particle growth from 1.4 nm to 100 nm.

……For example, the sensitivity of P_{theo} for particle growth from 1.4 nm to 100 nm is 12.0 at CS/GR = 50 nm^{-1}, indicating that with a typical 100% uncertainty in the measured GR, the uncertainty in P_{theo} can be as high as 12 orders of magnitude.

Authors cite a limited number of studies reporting the measured survival probabilities retrieved from aerosol size distributions (Page 2, Lines 50-53). Undoubtedly, the major limitation is validating the estimated survival probability for specific particle size (>1 nm) as it is not possible to track individual particles in the atmosphere and therefore their survival probabilities. Given this limitation, authors should consider referring to other methods/studies based on measurements and modelling approaches (typically different environments and therefore aerosol size distribution properties) such as Westervelt et al., 2013, Zhu et al., 2020; Sebastian et al., 2021; Pierce et al., 2014, etc., and survival probabilities for same particle size could be compared, discussed, and tabulated, which would make the reader easier to visualize.

Response: We added a new figure to the Appendix to summarize both P_{theo} and P_{meas} in literature. Although there were very limited studies comparing P_{theo} and P_{meas}, the literature values suggest that P_{meas} spread over a wide range even for the same type of environment and it seems to be higher than P_{theo}, especially for sub-3 nm particles.

Revised manuscript: On one hand, theoretical survival probabilities predicted using GR and the coagulation sink (CoagS) of new particles (Kerminen and Kulmala, 2002; Lehtinen et al., 2007; Pierce and Adams, 2007) have been widely used in regional and global models… On the other hand, however, there have been only a limited number of studies reporting the measured survival probabilities retrieved from aerosol size distributions (e.g., Weber et al., 1997; Kuang et al., 2009; Kulmala et al., 2017; Zhu et al., 2021; Sebastian et al., 2021). As summarized in the Appendix, only few studies have compared the measured and theoretical survival probabilities and the reported results seem to indicate that the measured survival probability is sometimes higher than theoretical values.

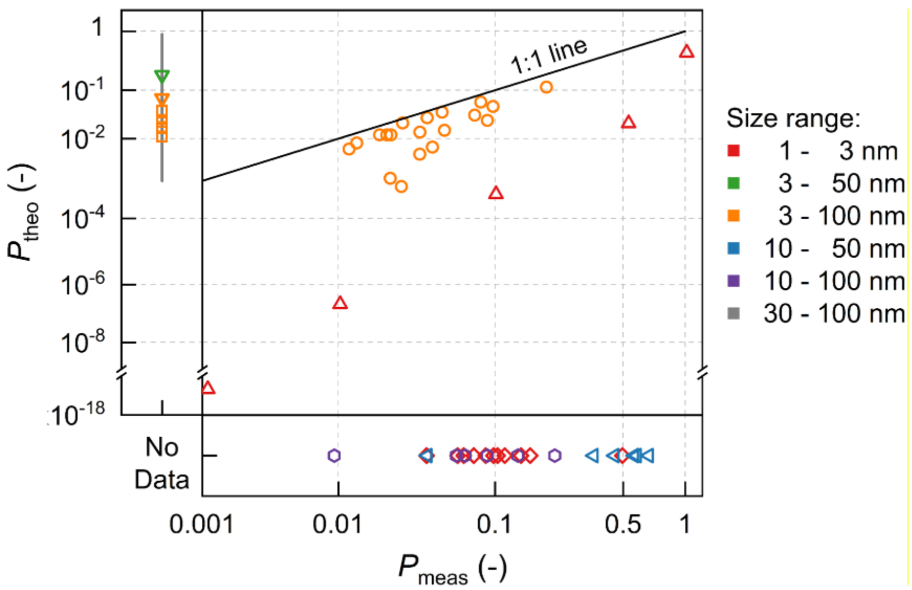

Figure A1: Particle survival probability in diverse environments. Data in this figure are collected from Weber et al. (1997), Pierce and Adams (2007), Kuang et al. (2009), Westervelt et al. (2013), Pierce et al. (2014), Kulmala et al. (2017), Zhu et al. (2021), Sebastian et al. (2021). Data from different studies are shown in markers with different shapes. Note that the axes are not on linear scales.

The regional NPF occurs in relatively homogenous air mass (the one shown in Fig 1a seems to have occurred in homogeneous air mass and can be checked by calculating air mass backward trajectories at each hour (0 to 24) for that day) so that atmospheric inhomogeneity can be avoided for NPF events to reduce underlying uncertainties. Any recommendation on how uncertainties arise from traffic emissions and other sources may be reduced/corrected?

Response: We added a case study of an NPF event (as also recommended by reviewer #2) to emphasize the unfavorable influence of atmospheric inhomogeneity on the measured survival probability. It may be difficult to correct the influences from traffic emissions and other sources on the measured survival probability because of the difficulty in distinguishing particles from different sources using the measured aerosol size distributions. There are some methods to correct the influences of transport and traffic emissions (Cai et al., 2018; Kontkanen et al., 2020), yet these methods are based on predetermined particle growth and loss rates, i.e., essentially the implicitly determined theoretical survival probability. Regarding these challenges, we recommend using statistical analysis to reduce the uncertainty related to atmospheric inhomogeneity, such as the average result shown in Fig. 6 in this manuscript and the scatter plots in other studies (e.g., Tuovinen et al., 2022). For case studies, we recommend checking the atmospheric homogeneity.

Revised manuscript: Figure A4 shows an NPF event measured at Hyytiälä as a case study for the significant influence of NPF on the measured aerosol size distributions. The measured mode dN/dlogd_p increased with a growing particle size until ~11:00, showing a high dN/dlogd_p region of new particles at ~10 nm. Consequently, P_{nlog} and P_n for 7-12 nm particles were larger than 1.0. Particle accumulation in a certain size range due to size-dependent particle growth rate was not the main cause of the high dN/dlogd_p region, as a clear pattern of rapid particle growth can be seen from the growing mode. The wind direction was relatively stable, though there was an increase in the wind speed at ~10:00. According to the analysis in Lampilahti et al. (2021), the high dN/dlogd_p region and therefore the unphysical values of P_{nlog} and P_n were likely to be caused by vertical transport of new particles. This vertical transport as an external source of particles is supported by the increasing total concentration of particles in the growing mode before 11:00. Interestingly, P_J coincides with P_{theo} though the value J computed using 25 nm as an upper size limit did not necessarily characterize the particle formation rate. For particles larger than 12 nm, the trend of P_{nlog} followed that of P_{theo} though there was still the influence of transport on the measured aerosol size distributions. This case study shows that for a certain NPF event, the measured survival probability may be heavily influenced by the inhomogeneity of the atmosphere. Analyses based on air homogeneity (e.g., backward trajectory), as well as statistical analyses based on long-term measurements, may help us to reduce uncertainties in measured survival probabilities.

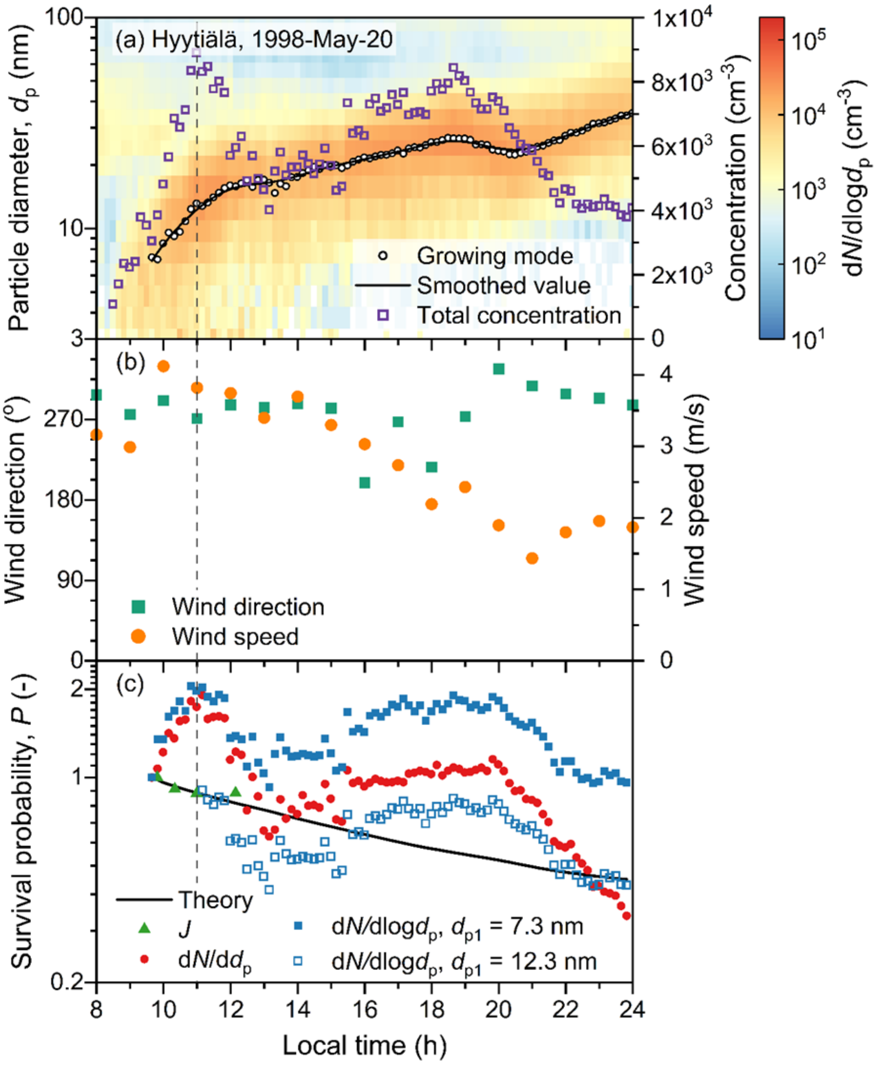

Figure A4. A case study of atmospheric inhomogeneity on the measured survival probability of new particles.

There are typos, not all but the critical ones can be taken care of. I list here a few of them

Page 2, Line 40: I feel it should read as “is an important irreplaceable parameter”

Page 2, Line 60: add “on” between “based” and “other parameters”

Page 3, Line 64: add “to” between “be equal” and “the ratio”

Page 8, Line 192: using “the” appearance time method

Page 4 and 8: “time-and-size-dependent”, it should be “time- and size-dependent”, or correct as appropriate throughout the text

Figure 7 caption: correct as “using the condensation sink (CS) “of” the sulfuric acid”

Response: Thanks. We have corrected these typos as well as those in other places in the manuscript.

Reviewer #2

The manuscript deals with evaluating possible approaches how to estimate particle survival property after nucleation events from measured particle size distribution dynamics. The approaches are tested by measurements in Hyytiälä and Beijing, representing growing nucleation modes after an event, and in the CLOUD chamber, representing approach to steady state dynamics. The analysis is very interesting, however, the number of studied cases is very limited in order to make the strong conclusions made, and one of the main conclusions may actually be incorrect (or at least misleading).

Response: We agree with the reviewer that the topic is interesting. In the revised manuscript, we clarify that the validity of different methods is guaranteed by benchmark simulations and the closure between average P_{theo} and P_{meas} is based on the statistics of the 1-year dataset. We also emphasize the uncertainties related to the influences of atmospheric inhomogeneity on P_{meas}. We understand the reviewer's concern that P_{nlog} may not valid for any growing aerosol population. This concern may originate from an ideal scenario that particle growth is driven by only non-condensable vapors, yet the growth of atmospheric particles is usually associated with other processes such as vapor dissociation. To address this concern, we added discussions and figures to clarify that the n_{log} method is based on a relatively constant geometric standard deviation (GSD). Such a constant GSD is an empirical conclusion for atmospheric new particle formation events and the n_{log} method in Eq. 7 has accounted for the variation of the GSD. Responses and discussions are given in detail below.

Major comments:

Perhaps the main comment of the manuscript is that n_{log} should be used to retrieve the measured survival probability for a growing aerosol population and n (or J) for quasi steady state distributions. My intuitive claim is that the choice should instead be made based on how GR depends on size. The authors' conclusion is based on a limited number of example cases where the nucleation mode width stays roughly the same, in log-scale, while the particles grow. Wouldn't this mean that GR should be (at least roughly) linearly dependent on size, so that for example 30 nm particles should grow 10 times faster than 3 nm particles? Is this always the case in the atmosphere? What if GR is roughly independent of size? Then the linear size distribution n should stay roughly constant in width (in linear scale) and the logarithmic one n_{log} should become narrower (in log-scale). For example, in Hyytiälä there are commonly particle formation events, where on the contour plot the most red color appears only after some growth (for example, May 20th, 1998), indicating that the n_{log} value increases while the mode grows. Then, obviously, n_{log} cannot be the choice for experimental survival rate estimation, as the result would be more than 100%. Thus, I urge the authors to analyze some events of this type also.

Response: The broadening of aerosol size distribution on the linear scale during growth does not necessarily require an increase in GR. The reviewer's intuition may be based on particle growth driven by the condensation of non-volatile vapors. As shown in Fig. A2 in the revised manuscript, there is no significant broadening on the linear scale and therefore using n_{log} without a GSD correction would significantly overestimate the survival probability. However, semi-volatile vapors may affect the size distributions of atmospheric particles. Fig. A3 shows that there is a significant broadening in n in the linear scale if the vapor can dissociate from particles, while the GR in Fig. A3 is kept the same as that in Fig. A2.

The evolution of GSD during particle growth is a synergistic result of coupled processes. For atmospheric NPF, there is usually a broadening of aerosol size distribution on the linear scale whereas the GSD can be relatively constant. We have also emphasized in the revised manuscript that Eq. 7, which accounts for the GSD, should be used if the GSD has a significant size dependency.

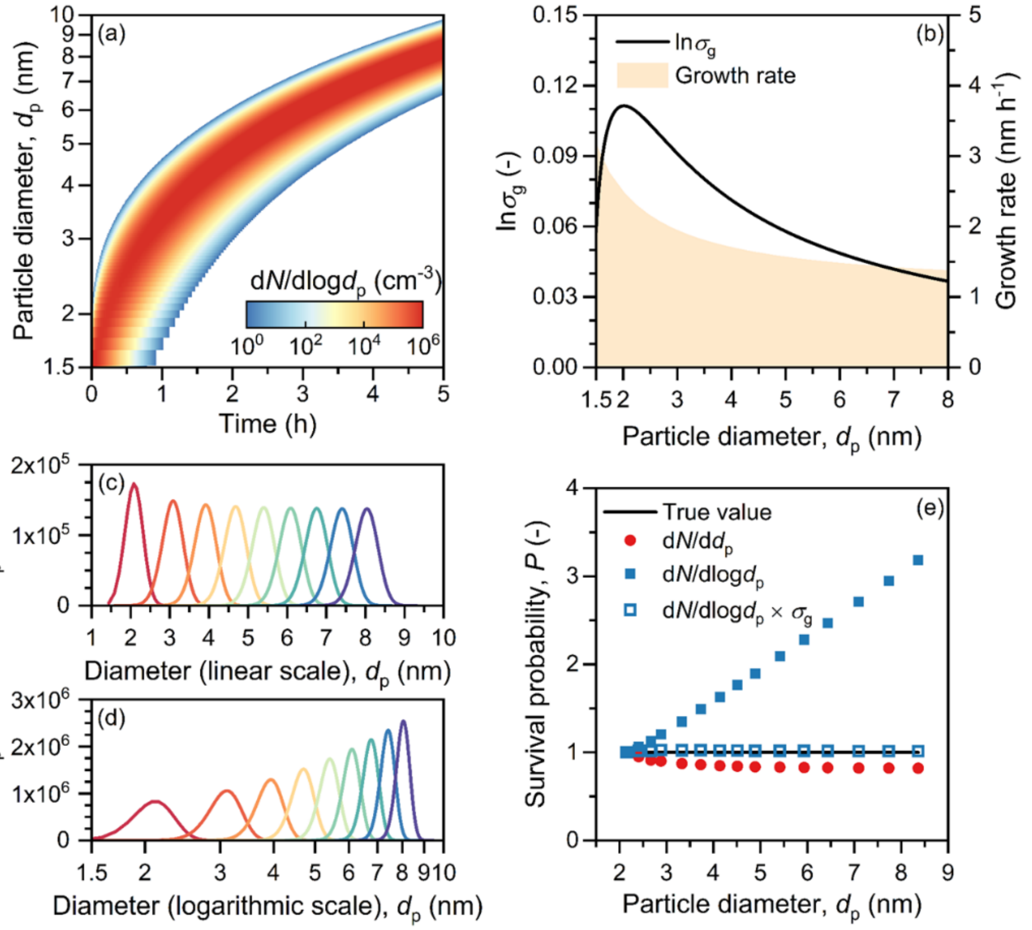

Figure A2: The growth of particles driven by the condensation of a non-volatile vapor

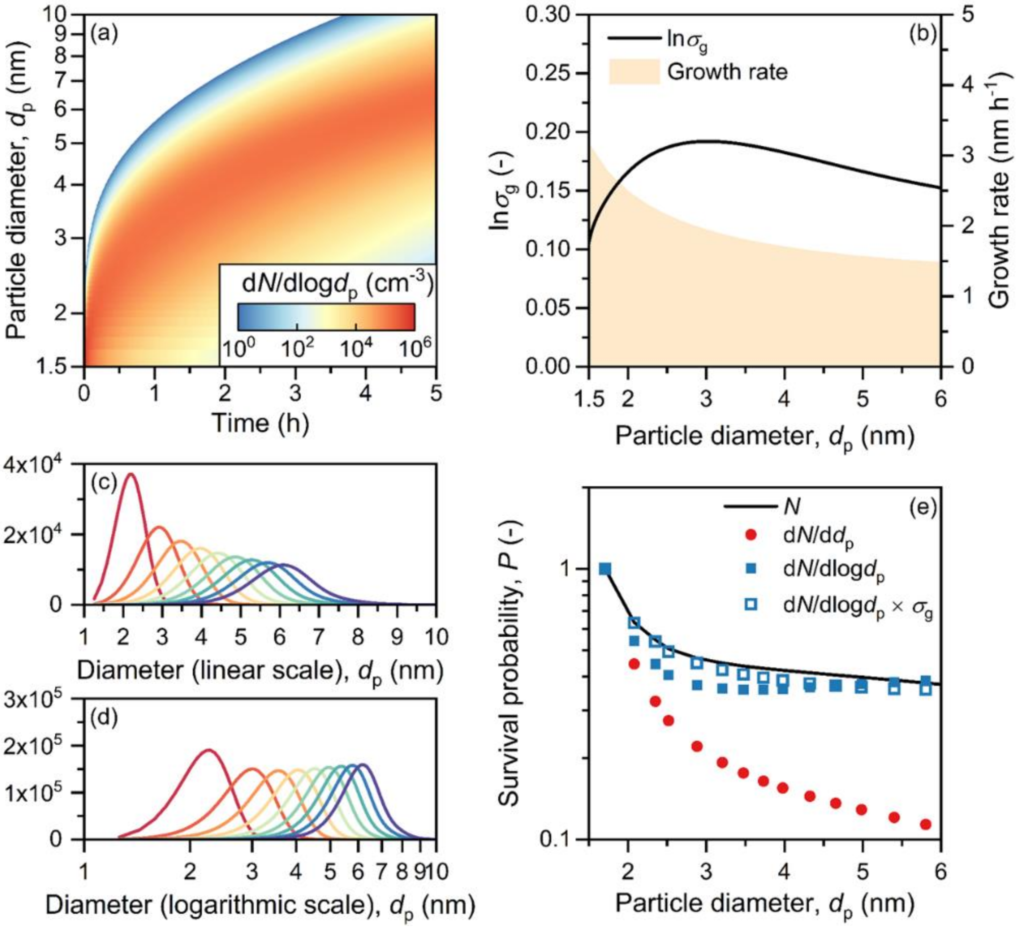

Figure A3: The growth of particles driven by the condensation of a volatile vapor and particle coagulation

We agree with the reviewer that high dN/dlogd_p regions are sometimes observed at large particle sizes and these events are common for the Hyytiälä site. We analyzed the event on May 20th, 1998, and use it as a case study to illustrate the uncertainties in P_{meas}. The evolution of GSD was accounted for when computing $P_{n\log}$. As shown in the figure below, $P_{n\log}$ for 7-12 nm particles (before 11 am) was higher than 100 %, indicating the failure of $P_{n\log}$ for this NPF event. However, this failure is not due to aerosol dynamics related to GR or GSD and the figure below shows that P_n was also higher than 100 %. This is also supported by the increasing concentration of nucleation mode particles and the large deviation between P_n and P_J (because the computed J characterizes the sum of formation and transport).

The most likely cause for this high dN/dlogd_p is vertical transport. Lampilahti et al. (2021) have shown that the downward flux of new particles from the residue layer to the mixing layer can have a significant influence on the aerosol size distributions measured in Hyytiälä. According to the analysis therein, NPF in the upper residue layer constitutes 42% of the NPF event days in Hyytiälä during 2013-2017. This case study provides good support for our argument that there may be variations in P_{meas} due to the complex inhomogeneous atmosphere. We added the analysis of this case study to the revised manuscript. We also recommend statistical analysis and checking the atmospheric homogeneity for case studies.

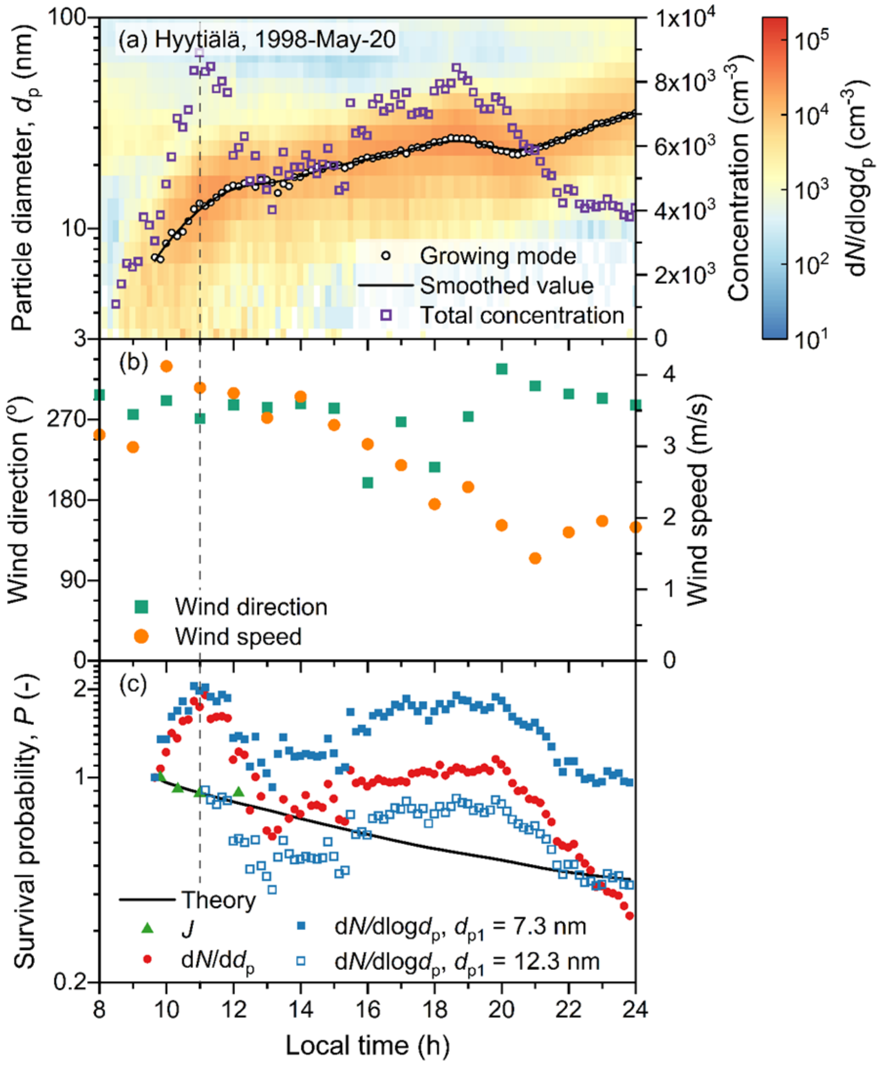

Figure 8. A case study of atmospheric inhomogeneity on the measured survival probability of new particles.

Revised manuscript: It is worth clarifying that σ_g may have a strong size dependency during some growth processes. For instance, if particle growth is only driven by the condensation of non-volatile vapors, σ_g tends to decrease as particles grow in size (see the Appendix). For those kinds of situations, Eq. 7 should be used instead of Eq. 6 for a better accuracy.

The influences of a size-dependent geometric standard deviation can be readily accounted for using Eq. 7.

Figure A4 shows an NPF event measured at Hyytiälä as a case study for the significant influence of NPF on the measured aerosol size distributions. The measured mode $dN/dlogd_p$ increased with a growing particle size until ~11:00, showing a high $dN/dlogd_p$ region of new particles at ~10 nm. Consequently, P_{nlog} and P_n for 7-12 nm particles were larger than 1.0. Particle accumulation in a certain size range due to size-dependent particle growth rate was not the main cause of the high $dN/dlogd_p$ region, as a clear pattern of rapid particle growth can be seen from the growing mode. The wind direction was relatively stable, though there was an increase in the wind speed at ~10:00. According to the analysis in Lampilahti et al. (2021), the high $dN/dlogd_p$ region and therefore the unphysical values of P_{nlog} and P_n were likely to be caused by vertical transport of new particles. This vertical transport as an external source of particles is supported by the increasing total concentration of particles in the growing mode before 11:00. Interestingly, P_J coincides with P_{theo} though the value J

computed using 25 nm as an upper size limit did not necessarily characterize the particle formation rate. For particles larger than 12 nm, the trend of P_{nlog} followed that of P_{theo} though there was still the influence of transport on the measured aerosol size distributions. This case study shows that for a certain NPF event, the measured survival probability may be heavily influenced by the inhomogeneity of the atmosphere. Analyses based on air homogeneity (e.g., backward trajectory), as well as statistical analyses based on long-term measurements, may help us to reduce uncertainties in measured survival probabilities.

Calling 'theory' the survival rate obtained by following the peak on a contour plot and integrating the competition between growth and scavenging along this 'trajectory' is a poor choice, as it is just another approximation. Size dependent scavenging causes apparent growth (see Leppä et al., ACP 11, p. 4939, 2011) and size dependent GR deformation of the size distribution shape, which means that the 'trajectory' obtained by following the peak of the nucleation mode (in log-space) might not represent the same aerosol particles. Please comment on this and if you agree, a change of terminology is needed.

Response: We agree with the reviewer that the evolution of the fitted mode peak is not the particle growth trajectory. Correspondingly, we revised "trajectory" as "mode diameters" throughout the manuscript and clarified that the "particle growth does not exactly follow the increasing mode diameters due to the influences of coagulation". For the simulated NPF in Fig. 4, we have used the growth trajectory of a single particle (instead of the mode diameter) to compute both P_{theo} and P_{meas}. For P_{theo}, the main uncertainty originates from the computation of GR, as we have discussed for sub-5 nm particles. Integrating CoagS/GR over the mode diameters is equivalent to accounting for the temporal evolution of CS, which does not affect the uncertainty related to GR computation.

Revised manuscript: For measured NPF events, the growth trajectory is approximated by the evolution of particle mode diameters, though particle growth does not exactly follow the increasing mode diameters due to the influences of coagulation (Stolzenburg et al., 2005; Leppä et al., 2011).

The P_{theo} for atmospheric new particles was computed using Eq. 2, with the CoagS determined along the mode diameters and the concentration of new particles numerically solved by iteration.

As GR is such an essential parameter when considering survival, please add figures showing the size-dependent GR (and time dependent also, if there is time-dependence) in the simulated cases. Now there is only a vague statement on page 7 (line 157) that "A growth enhancement factor for particle growth (Kuang, 2010) was used....."

Response: We thank the reviewer for this constructive suggestion. The growth rate in the simulated cases has been added to Figs. 2-4 and A2-A3. We also clarify that the growth rate in Fig. 4 is time- and size-dependent and we show the growth rate along the growth trajectory.

Revised Figure 2 (The growth rate can be seen as the shaded area in panel b):

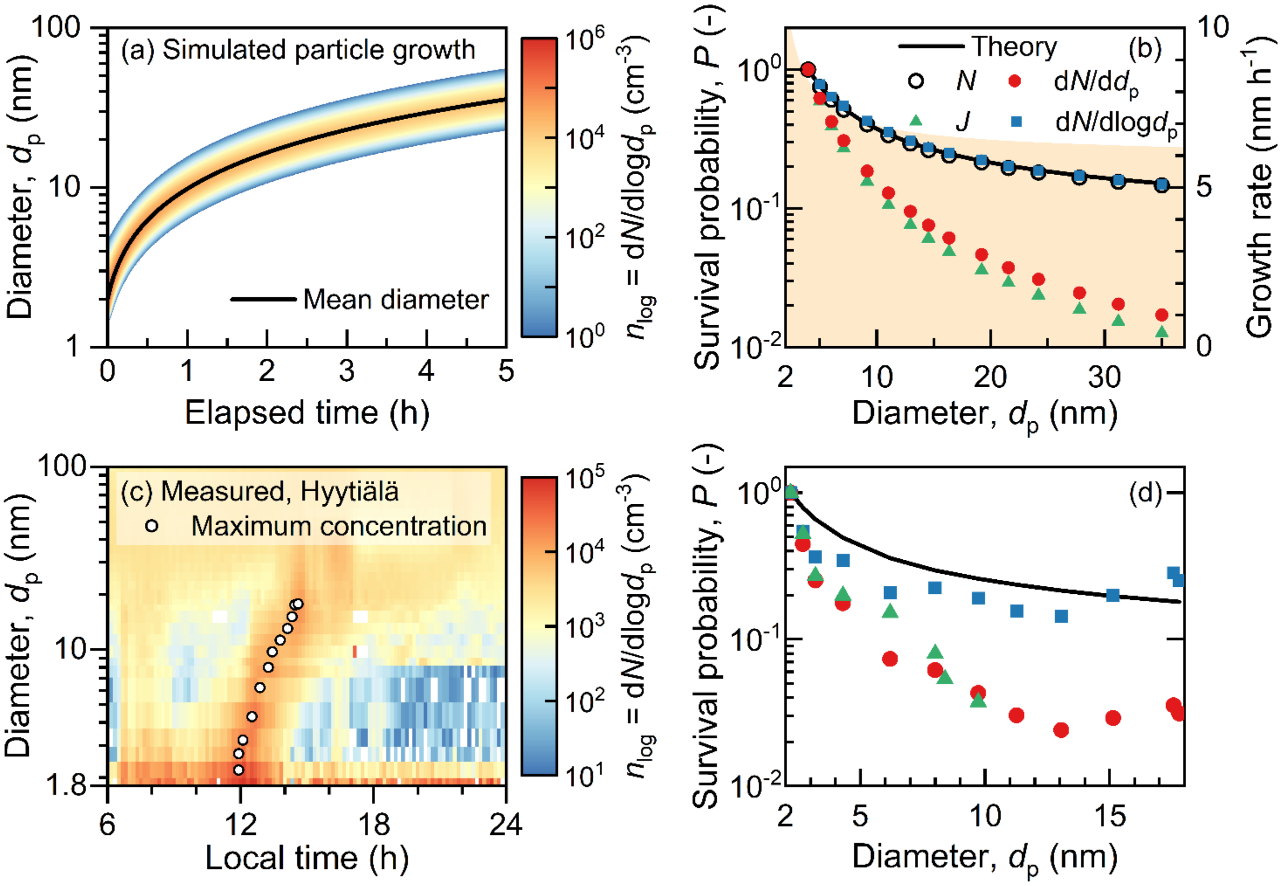

Figure 2: Survival probabilities of new particles in simulated and measured growing aerosol populations.

One puzzling observation has been observed in polluted megacities such as Beijing: how can the particles survive with such high sink-values and low growth rates? The authors now claim that this proposed way of estimating survival rate, based on using n_{log}, resolves this issue. This is an important, intriguing question, which is discussed here quite loosely, especially since many of the authors have another manuscript being reviewed at the same time on this specific topic (Tuovinen et al., ACPD). Much more impressive would be to use full simulations by the sectional model that the authors have in their use, with observed sink and GR values to see if the nucleated particles actually survive - this may be, however, a topic for another publication. Now it remains a bit unclear, based on reading this manuscript alone, what is really the conclusion regarding analysis of the events in Beijing.

Response: We agree with the reviewer that modelling the aerosol size distribution and comparing it with measurements will better clarify the survival of particles in Beijing. We have an ongoing study that will address this problem based on the comparison between modeling and measurements.

We would like to clarify that we did not use the n_{log} method to resolve the puzzle of particle survival in Beijing. As has been clarified in Section 4.2 and Fig. 6, P_J was used for sub-10 nm particles and P_n was used for sub-25 nm particles for NPF events in urban Beijing. We also clarify in the revised manuscript that despite the advances in this study and related studies, the survival of sub-3 nm particles under high CoagS in Beijing is still a puzzle.

Revised manuscript: Note that P_J is used for sub-10 nm particles, which is consistent with the method in previous studies.

However, NPF events can occasionally be observed under high CoagS and there are deviations between the P_J and P_{theo} of sub-3 nm particles during these events (Tuovinen et al., 2022).

Minor comments:

6. Equation 2 and related text: what is *N* actually for a continuous distribution? It is clear what it means for a monodisperse one, but if one wishes to follow survival rate for a 'real distribution', shouldn't *N* be then the total number concentration for some size interval?

Response: We clarified that *N* is herein the total concentration of the growing mode particles. This growing mode may be a hypothetical population, e.g., as a subset of the overall distribution for which particles shares the same initial size. As long as one can separate particles in this growing mode from other particles, e.g., in a model or in theoretical derivations, it can be unnecessary to define a certain size interval.

Revised manuscript: where *N* is the total concentration of particles in the growing population.

7. Page 5, lines 123-124: It is claimed that the GSD usually remains relatively constant for atmospheric particle formation events. Is there a reference supporting this? As mentioned in comment 1, there are several events in Hyytiälä at least, where the peak value of $n_{\log}$ increases along with growth, indicating simultaneous narrowing of the growing mode also.

Response: We added a reference reporting the GSD to support this empirical conclusion. There can be cases where the GSD evolves with particle size, hence we start from Eq. 7 which accounts for the evolution of the GSD and clarify that the GSD may have a strong size-dependency during some growth processes.

Eq. 7 is used to analyze this NPF event in Hyytiälä. According to the discussions above, $P_{n\log}$ and P_n were higher than 100 % for 7-12 nm particles because the atmospheric homogeneity assumption was not valid.

Revised manuscript: ……it is an empirical conclusion that σ_g usually maintains a relatively constant level (e.g., Hussein et al., 2004).

"It is worth clarifying that σ_g may have a strong size dependency during some growth processes. For instance, if particle growth is only driven by the condensation of non-volatile particles, σ_g tends to decrease as particles grow (see the Appendix). For these growth processes, Eq. 7 should be used instead of Eq. 6 for better accuracy."

8. Page 8, line 187: Explain in detail how the growth trajectories were obtained. Are they based on peak values in log-scale? Has smoothing or fitting been used? If yes, please state the details.

Response: We present the details for obtaining the mode diameters and maximum concentrations. The "growth trajectory" was no longer used for measured NPF events, as the trend of mode diameters is not exactly the growth trajectory.

Revised manuscript: For simulated NPF events, the growth trajectory was obtained using a monodisperse aerosol model. For measured NPF events, the growth trajectory is approximated using the evolution of particle mode diameters or the maximum concentration method, though particle growth does not exactly follow the increasing diameters due to the influences of coagulation (Stolzenburg et al., 2005; Leppä et al., 2011).

The mode fitting method tracks the growth of peak diameter of a new particle mode by fitting lognormal distributions to the measured $n_{\log}$.

For Hyytiälä, the GR was retrieved using the maximum concentration method (Kulmala et al., 2012), which finds the time corresponding to the maximum particle concentration in each size bin, and the concentration was smoothed with a span of 12 min.

9. Page 8, lines 205-206: It is stated that the used *J* is the daily maximum for each size bin. What does this mean?

Response: We determined the temporal evolution of $J(d_p)$ and took the maximum value to represent the growth flux of particles. That is, *J* was not determined along the mode diameters. According to our simulation results, this method is more robust than using *J* along the mode diameters.

Revised manuscript: ……the *J* in Eq. 4 as the maximum *J* during an NPF event as a function of particle size.

10. page 9, line 220: The definition of equation 1 is very clear for a monodisperse growing mode, but as I explain in my comment #2, it is unclear how growth of the "same population" can be determined from a continuous evolving distribution.

Response: Eq. 1 is mostly a definition equation, which means it may not be straightforward in applications. For the real atmosphere, it is difficult to determine whether a particle belongs to this population or not without prior knowledge. We have clarified in section 4.1 that "Due to the continuous particle formation, it is difficult to apply the definition of survival probability in Eq. 1 to the simulated NPF. Misapplying Eq. 1 by taking all the measured particles as survived particles would result in survival probability values larger than unity (Fig. 3b)."

11. Figures 2a and 4a, and respective simulations: Is the relatively constant width of the growing mode obtained by setting an appropriate size dependence of GR on d_p see also comment 1), or is there also some numerical diffusion present?

Response: We are aware that there is indeed unavoidable numerical diffusion in the simulation results. However, we have clarified that "We validated the accuracy of the sectional model using a discrete model, ensuring that numerical diffusion did not affect the conclusions based on simulation results." Simulation results using a discrete model are given in Fig. A2-A3, which explains that the broadening of aerosol size distribution during particle growth by condensation is possible. We did not set the size-dependency intentionally to obtain a relatively constant width. The revised Fig. 2b shows a GR with a weak size dependency and Fig. 4c shows a GR with strong a size dependency.

12. Finally, if possible, the authors could discuss more what is actually a 'true' survival probability. All methods presented here are approximations, even the one that is called 'theory' in this manuscript (comment 2). Somehow, intuitively, if there is only condensational growth and scavenging, it should be the survival probability of a size interval of particles, that obviously stretches (or gets narrower) in 'length' if there is size dependent GR. Also J, intuitively, should be one obvious candidate. This is why the results of this manuscript are so interesting, showing that in many cases experimental n_log seems to work quite well (if the used trajectory-based analysis as a comparison is accepted as a valid one).

Response: We are a bit confused by the comment on the "true" survival probability. The true survival probability is defined with respect to individual particles. An equivalent definition of the true survival probability is given in Eq. 1, and we use it as a benchmark in Figs. 2 and A3.

We agree that both the theoretical and measured survival probabilities are approximations. We also agree with the reviewer that GR and GSD may affect the aerosol size distribution, so we have included GR in Eq. 11 and GSD (as a result of GR and other causes) in Eq. 7.

We agree that *J* is an obvious candidate. We demonstrate the validity of *J* for pseudo-steady-state aerosol size distributions. However, we have also shown that the *J* method may lead to a bias for a growing aerosol population.

It seems that the reviewer's major concern is on the relatively constant GSD of aerosol size distributions during particle growth, which is against the intuition that there should be no broadening in the linear scale with a constant GR. Such a concern may also be related to some statements in the original manuscript. To address this concern, we show in Fig. A3 above that broadening in the linear scale can be a natural result of particle growth. We also clarify in the revised manuscript that Eq. 7 should be used instead of Eq. 6 if there is a significant size-dependency of GSD.

References

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