Responses to Reviewer #1's Comments on Manuscript acp-2020-398

(Impacts of coagulation on the appearance time method for sub-3nm particle growth rate evaluation and their corrections)

We thank Prof. Dr. Wolfgang Junkermann (referred as reviewer #1 below) for the efforts and constructive comments that help to improve this manuscript. The reviewer's comments are addressed in the following paragraphs and the manuscript were revised majorly. In response to the concerns of reviewer #1, we clarify in the revised section 3.1 that the numerical models in this study are used to provide a benchmark to test the proposed formulae rather than investigate the nucleation mechanism in the real atmosphere. The comments are shown as sans-serif dark red 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. Line numbers, figures, and equations quoted in the responses correspond the revised manuscript. References are given at the end of the responses.

Reviewer #1

Summary

The manuscript describes a numerical set of formulas that is used in an aerosol model for the calculation of the growth of aerosol particles after nucleation, especially for the range of ~ 1 nm to 3 nm. This formula set is used for a calculation of the appearance time (time after cluster formation) and later for estimation of the growth rate of the larger sizes aerosols > 4 nm although the main emphasis is on the smallest size ranges, where coagulation has a major effect. Model results based on these formulas are finally compared to a nucleation mode particle event in a relative highly polluted environment in China, in the city of Beijing in a late winter situation.

Response:

This manuscript describes the formulae to correct the influence of coagulation on the appearance time method for growth rate estimation, especially for the range of ~1-3 nm. The derivations of the conventional and corrected appearance time methods for growth rate estimation are detailed in Sections 4.1 and 4.2. In addition to derivations, we also test these formulae for the conventional and corrected appearance time methods using aerosol dynamic models (Section 4.1-4.3, Figs. 1-6). The comparison between the growth rate given by the model and the rates retrieved for the simulated evolution of aerosol size distribution using the conventional and appearance time method support the argument that the impact of coagulation should be corrected when using the appearance time method to estimate the new particle growth rate. Further, we provide an example to illustrate the effect of coagulation on the growth rate retrieved using the appearance time method in the real atmosphere in urban Beijing. A comparison between the growth rates retrieved using the conventional and corrected appearance time method is shown during a new particle formation event in Beijing. There is no comparison between the measured size distributions in urban Beijing and the simulated distributions using the aerosol dynamics models in this study. These research methods are also summarized in the last paragraph of the Introduction section.

According to the reviewer's comments below, a majority of the reviewer's concerns are closely related to the comment "Model results based on these formulas are finally compared to a nucleation mode particle event". Detailed responses will be given following every comment and we present a summary of these responses in the following paragraph.

In the revised manuscript, we clarified that models used in this study are based on aerosol dynamics but not the formulae for conventional or corrected appearance time methods. A paragraph is added at the beginning of Section 3 for

a better understanding. The input of the models are an initial aerosol size distribution and the concentration of the gaseous precursor as a function of time. The evolution of the aerosol size distribution is obtained by solving the aerosol general dynamic equations. Meanwhile, particle growth rate as a function of particle size and time is given by the aerosol dynamic model, which is calculated according to the condensation, evaporation, and coagulation rates. The formulae for conventional or corrected appearance time methods are used to retrieve particle growth rate from the simulated evolution of the aerosol size distribution. Since the simulated growth rate (given by the aerosol dynamic model) is consistent with the simulated evolution of the aerosol size distribution while there are approximations in the estimation using the appearance time methods, the comparison between the simulated growth rate and the growth rate retrieved using the appearance time methods indicates the uncertainty of the appearance time methods in the simulation conditions. The simulation conditions are chosen to elaborate not only the accurateness and uncertainties of the appearance time methods but also the causes of these uncertainties. For instance, Fig. 5 shown the accuracy of the corrected appearance time method under a constant vapor concentration, which support the derivations for the appearance time method. However, since the derivations are based on a constant vapor concentration assumption whereas the vapor concentration in real new particle formation events may vary with time, we present Fig. 6 and Table A1 to shown the uncertainties due to this violation of the constant vapor assumption. After testing the appearance time methods using the aerosol dynamics models, the appearance time methods are applied in a new particle formation event measured in urban Beijing. The growth rate during this event are estimated using the conventional and corrected appearance time methods and their differences indicate the influences of the coagulation on the appearance time of new particles. The measured size distributions and the retrieved growth rates for this new particle formation event are not compared to those given by the aerosol dynamic models.

We revised the first sentence in Section 3.1 as "A discrete aerosol model and a discrete-sectional aerosol model based on aerosol dynamics were used to provide an evolving aerosol size distribution and hence to test the conventional and corrected appearance time methods" and added "Note that the aerosol dynamic models are only used to provide a benchmark to compare the conventional and corrected appearance time methods in this study" at the end of this section.

General comments

The argumentation and description of the formulas seems plausible and is according to the authors in agreement with the theory. Unfortunately this 'reference' theory, which is several times cited in the manuscript, is neither described nor referenced. Also there is no information about this general 'theory' and the underlying nucleation scheme applied. There are several nucleation schemes, binary nucleation of sulfuric acid and water (neutral and ion-assisted), ternary involving sulfuric acid, water, and ammonia; with or without organic species and sulfuric acid or charged sulfuric acid-water-ammonia (Napari et al, 2002; Riccobono et al. 2014; Dunne et al. 2016). Otherwise, the results of the subsequent calculations are at least partially contradicting previous publications that are a base for many 'new particle formation' studies within the last decade.

That discrepancy is likely due to the fact that the calculations are done only with a limited set of condensing substances, likely H_2SO_4. It is well known that the exclusive condensation of H_2SO_4 is too slow for the growth rates observed in the atmosphere and that other substances like VOC's or ELVOC's have a large share on growth rates (Ehn et al, 2014; Kuang et al, 2012; Kupc et al, 2020 and literature cited there). An overview about the problems associated with the investigation of nucleation under Chinese high pollution conditions is given by Chu et al (2019). Here also potential other compounds that might be important for NPF. Cai et al (2017) showed that H_2SO_4 could and should be involved but, high H_2SO_4 concentrations do not necessary lead to particle formation events.

Response: We agree with the reviewer that the nucleation and condensation of a single vapor (e.g., H_2SO_4) is usually insufficient to explain new particle formation in the atmosphere. However, the aim of this study is to investigate how to

estimate the growth rate from the measured aerosol size distributions rather than figure out the compounds contribute to particle growth and the their growth mechanisms. The aerosol dynamic models used to test the formulae of the appearance time methods and they are not used to illustrate the nucleation and growth mechanism. Hence, the simplifications in the aerosol dynamics models in terms of the nucleation mechanism does not affect the tests because they do not affect the consistency between the growth rate and evolution of the aerosol size distribution given by the model. There is no modeling results in the application example in Fig. 7.

A new paragraph was added at the beginning of Section 3 to clarify the reason to use models.

Although it does not influence the statement of the reviewer in this piece of comment, we would like to briefly clarify the conclusion of a previous study (Cai et al., 2017). The reason that high H_2SO_4 concentrations do not necessary lead to particle formation events in urban Beijing is usually because of the aerosol Fuchs surface area concentration (coagulation sink of new particles). When H_2SO_4 concentration was high and coagulation sink was low, there were usually new particles observed in urban Beijing. However, this does not indicate that H_2SO_4 is the only major compound participate new particle formation in urban Beijing and the contributions of other compounds to particle growth were discussion in Cai et al. (2017).

The physical background besides the description of the mathematical steps is largely missing. Neither the environmental conditions for the validity of the model, nor the initial input parameters, temperatures humidity, number and size of large particles acting as a condensation sink etc. are included. 100 nm particles are mentioned in a 'certain' number.

Response: To clarify the background particles used in the aerosol dynamic model, we added "The condensation sink (10^{-3} – 10^{-2} s^{-1}) is contributed simultaneously by a certain number concentration (7.2×10^{2} – 7.2×10^{3} cm^{-3}) of 100 nm background particles and the new particles." Other input parameters (e.g., temperature) are omitted because they does not affect the mathematical derivations and the aerosol dynamics models are only used to test these derivations in this study.

The major difference between the new approach of the 'appearance time method' and the 'old' theory, that needs to be discussed in detail is the result that, for the small particles size the growth rate due to coagulation is slowed down compared to other studies where the growth rate is continuously increasing with size (Kulmala et al, 2004, Kulmala et al, 2013, Kuang et al, 2012). The result is a longer appearance time (12 hour for 4.9 nm) than published elsewhere (Kulmala et al, 2013). For a detailed description of the current understanding of the initial steps of gas to particle conversion, and probably the 'theory' mentioned in the manuscript see also Kulmala et al, 2017 and Yao et al, 2018.

Response: We agree with the reviewer that correcting the effect of coagulation reduces the value of growth rate retrieved using the appearance time method. However, there is no discrepancy of the size dependency of growth rate reported in this study and in previous studies. Figure 7b shows that the growth rate estimated using the corrected appearance time method generally increase with the particle size, while the slightly negative size dependency in the sub-3 nm size range is mainly due to the decreasing H_2SO_4 concentration and the size dependency of the coagulation coefficient.

The apparent relationship between particle growth rate and particle size does not necessarily characterized the size dependency of particle growth rate. In the revised section 2.3, we clarified that "Note that since the appearance time is a function of d_p and GR_{conv} is estimated for difference appearance time, the apparent relationship between GR_{conv} and d_p does not necessarily indicate the size dependency of GR_{conv} at any given moment." For example, the methods in Kulmala et al. (2004, 2013) and this study only provides the growth rate as a function of both particle diameter and its corresponding appearance time. One has to exclude the influence of time-dependent variable (e.g., the varying vapor

concentration) to obtain the size dependency of growth rate. In contrast, Kuang et al. (2012) estimates the size dependency of particle growth rate using a time-and-size-resolved method via solving the aerosol general dynamic equation.

The "longer appearance time (12 hour for 4.9 nm)" refers to Fig. 6, in which the model was used to test the formulae. Hence, the simulated aerosol size distributions in this figure should not be directly compared to the measured size distributions in an atmospheric new particle formation event. Further, the appearance time corresponding to a particle size, by its definition, is a moment rather than a duration. We revised "12 h" as "12:00" to avoid confusion. As shown in Fig. 6, the duration for a particle to grow from 1.0 nm to 4.9 nm is ~2.5 h, which is comparable to the values in previous studies. However, this value is mainly determined by the input H_2SO_4 concentration and it has little contribution to understanding the nucleation mechanism because the models in this study are only used to test the formulae.

The model results are then compared to an event 'typical' for Beijing, with nucleation mode particles observed in late winter (24.2.2018). Also, in this section the authors do not give any details about the environmental conditions at the day of the particle event, nor any detailed information about the location, distance to major roads, height of the aerosol inlet above ground etc.. This information is important in a study about aerosols that may be affected by strong local sources (traffic, home emissions) and potential regional transport. Included is only a reference to a submitted, but still not yet accepted, companion paper. Also, as this is an aerosol study under conditions with a large condensational sink, it would be helpful to have data on fine particle concentration, a 'certain number of 100 nm particles' is way too uncertain.

Response: The example of the new particle formation event in urban Beijing is used to elaborate that under the relatively high coagulation sink in urban Beijing, the influence of coagulation on the appearance time of new particles and hence the retrieved growth rate is non-negligible for sub-3 nm particles. The growth rate estimated using the appearance time method in Fig. 7b was calculated using the measured aerosol size distributions. The measured size distributions in Fig. 7a were not compared to the simulated distributions in Figs. 1-6.

To clarify the environment conditions during this new particle formation event, we added "During 8:00 – 16:00 on this NPF day, the mean temperature, relative humidity, and wind speed are 1.3 °C, 22%, and 1.4 m s^{-1}, respectively" in section 3.2. A recently published manuscript, Deng et al. (2020b), reported the data measured at the same site during the same period. This reference was included to provide detailed information on the measurement site.

The measured aerosol size distribution ranging from 1-400 nm are shown in Fig. 7a. Note that "a certain number of 100 nm" particle were only used in the simulation shown in Figs. 3b – 6, and the simulation was not compared to measurements.

The most important issue, however, is the classification of the experimental comparison data. It is the timing of the event that makes it difficult to match the event with basic atmospheric physics and chemistry and with the current observations that the majority of NPF events are happening during daylight hours. A new particle formation event at that time of the day would require an even faster growth than shown by Kulmala (2013) and not a slower growth as indicated in this manuscript. Both, the 'old' Kulmala 4 hours and the 'new' 12 hours appearance time require nighttime chemistry and physics which has not been reported as a significant source for nucleation.

Response: We have revised "12 h" as "12:00". As shown in Fig. 7a, it took ~2 h for particles at 1.5 nm to grow up to 5 nm during the event on Feb. 24, 2018.

The data presented rather indicate a transport related change of air masses. The H_2SO_4 concentration which is at background levels at sunrise peaks about one hour after sunrise and then steadily declines throughout the day (Fig. 6 of the manuscript). This behavior is contrary to the model where the peak concentration is 4 hours later at noon. This experimental diurnal pattern is neither in agreement with local emissions in a high NO_x environment nor with a photochemical reaction including OH radicals and SO_2.

Response: The H_2SO_4 concentration used in the model (Fig. 6) was an input parameter because the model was used only the test the appearance time method rather than predict the H_2SO_4 concentration. It was determined to be either constant or follow a normal distribution to investigate the influence of a varying vapor concentration on the appearance time method, as detailed in section 4.3.

We agree with the reviewer that transport, mixing, and local emissions may influence the measured H_2SO_4 concentration. However, due to the high condensation sink and hence short atmospheric residence time of H_2SO_4 in the presence of a relatively high aerosol concentration, the measured H_2SO_4 is more likely to form at the observation site than being transported from elsewhere. Figure R1 shows the diurnal variation of SO_2 concentration on Feb. 24, 2018. The steadily decreasing H_2SO_4 concentration in Fig. 7 is in accordance with the decreasing SO_2 concentration between 8:00-16:00, indicating that the photochemical reaction between OH radicals and SO_2 is a major pathway for the formation of H_2SO_4 during this new particle formation event.

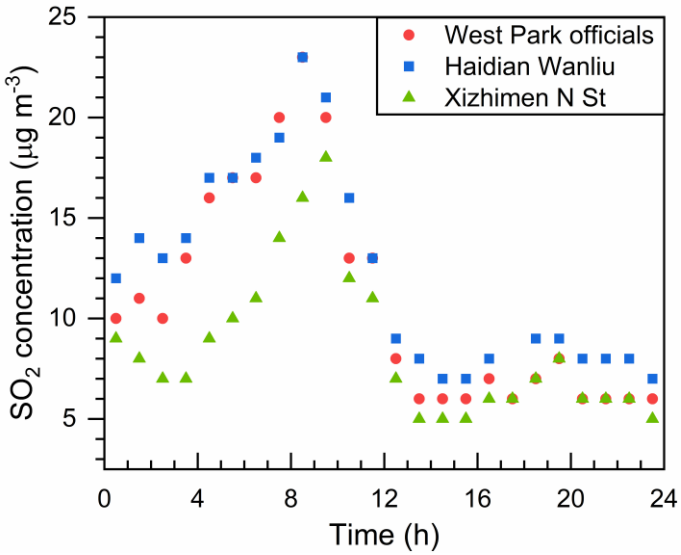

Figure R1. SO_2 concentration measured at three nearest national monitoring stations on Feb. 24, 2018.

Following Kulmala et al (2013) the appearance of 5 nm particles needs about 4 hours after initial cluster formation. This implies for the Beijing case study that cluster formation would occur before sunrise. The air mass at that time has been, according to HYSPLIT, in the area close to the city of Tangshan, not locally in Beijing. Following the results of the current study (appearance time of 4.9 nm particles ~12 hours) clusters would originate even further to the east, close to the coastline at a formation time a few hours after sunset (Fig. 1). This requires a discussion of a nighttime cluster formation chemistry.

Response: As clarified above, Fig. 7a shows that during this new particle formation event, the appearance time for 1.5 nm and 5 nm aerosols are approximately 8:00 and 10:00, respectively. As clarified above, the 12-h growth is a misinterpretation of this manuscript. This appearance time and the measured aerosol size distribution show that new particles down to the cluster size were observed at the site after sunrise and they grew steadily into large particles. Such

a complete formation and growth pattern of new particles indicates a typical regional new particle formation event (Kulmala et al., 2012), although transport, mixing, and local emissions may influence the measured aerosol size distributions.

Along the back trajectories several anthropogenic sources for sulphur compounds are located, that emit a mixture of both, particles and gas phase precursors (Fig. 1). Their emissions include sulfur dioxide, H_2SO_4 (Srivastava et al, 2004) and large amounts of ammonia (Li et al, 2017) and primary nanoparticles produced at elevated temperatures and with or without catalysts, Bai et al, 1992). These are than released into elevated layers of the atmosphere (Junkermann et al, 2011). Measured emission rates of such 'new aerosol generators' are in the order of ~3*10^15 s^{-1} MW^{-1}. Naturally, the emitted particles undergo coagulation and further growth or shrinking (evaporation) depending on the ambient conditions (temperature, humidity) during transport under cool and dry nighttime conditions and the plume conditions favor additional gas to particle conversion (Mohnen and Lodge, 1969). The shape of the diurnal pattern of H_2SO_4 in Beijing is in good agreement with such an advection-convection driven transport, by far better than with a local chemistry initiated cluster formation.

The example of the early morning particle event in Beijing is probably a typical 'nanoparticle' but not a formation event. The experimental verification is looking reasonable but likely does not reflect reality.

Response: We added a paragraph in Section 4.4 on the classification of the observed new particle formation event:

"As shown in Fig. 7, the measured H_2SO_4 concentration started to increase around 7:00 and its 50% appearance time was ~7:30. Because SO_2 concentration declined between 8:00 and 14:00, the peak concentration of H_2SO_4 was observed at 8:00 rather than at noon. Shortly after the increase in H_2SO_4 concentration, new particles down to the cluster size (~1 nm in geometric diameter) was observed. The high concentration of sub-2 nm aerosol during 7:00 - 15:00 and the continuous growth pattern of new particles from 1.5 nm to 10 nm indicates that the observed new particle formation event was a typical regional event (Kulmala et al., 2012), though transport, mixing, and emissions might influence the observed aerosol size distributions."

We disagree with the reviewer's hypothesis that the measured H_2SO_4 and aerosol size distribution at the site were predominated by transport. Due to the short atmospheric residence time of H_2SO_4 and sub-3 nm aerosols (Deng et al, 2020b), it is unlikely that they had been transported from somewhere 150 km away before the detection.

The agreement between the trends of the H_2SO_4 concentration in Fig. 7a and the SO_2 concentration in Fig. R1 supports the hypothesis that the daytime H_2SO_4 during this event was mainly formed by photochemical reaction. The continuous formation and growth pattern of particles down to the cluster size (~1 nm in geometric diameter) shown in Fig. 7a indicates the observed new particle formation event was a typical regional event. Furthermore, the agreement between the 50% appearance time of H_2SO_4 (7:30) and 1.5 nm particles (8:00) in Fig. 7a indicates that the H_2SO_4 formed after sunrise initiates the observed new particle formation event.

The manuscript would need a complete revision due to the missing model and experiment description and not verified nor discussed assumptions about the classification of the particle event. It's also required to discuss the differences between the current model and the previously published growth rate versus particle sizes and the implications on the timing of the whole nucleation process (cluster formation at night versus photochemical reaction).

The authors also should make sure that the appearance of nucleation mode particles in Beijing is applied to a really local phenomena and not to a plume study.

Response: Here we present a summary of revisions in response to the major concerns in the general comments of the reviewer.

- We clarified that the models in this study were used to test the proposed formulae for the appearance time method. There is no comparison between the modeling results and measurements in this study. Besides, we clarified the input background aerosol concentration for the model in the revised manuscript.
- We added more details on the experiments.
- We added a paragraph on the classification of the new particle formation event shown in Fig.7. The continuous formation of aerosols down to the cluster size and their subsequent growth are in accordance with the features of a regional new particle formation event rather than those of a plume event.
- The size-dependency of particle growth rate in the observed new particle formation event is similar to those reported in previous studies.
- The appearance time of H_2SO_4 and new particles on Feb. 24, 2018 indicates that the observed intensive new particle formation was mainly driven by daytime photochemical reaction.

Specific comments:

Model output

The model based on the new formula predicts initially a slow down of the growth rates of new particles within the size range from 1 to 3 nm. The behavior of the growth rates in the 'new' model setup of the manuscript would delay the growth of newly formed particles significantly compared growth rates published elsewhere (Kulmala et al, 2004 and Kulmala et al, 2013). In these publications a continuously increasing growth rate is used. Accordingly, with the new model the growth of new particles to sizes of about 5 nm takes ~10-12 hours compared to 4 hours in the Kulmala et al (2013) example. This is a severe difference likely due to the restriction of only one condensing vapor and has to be discussed in detail because it has strong implications for the time and location of the cluster formation process.

This difference implies several questions that are important for the model – experiment comparison (probably not for the calculation of the impact of coagulation but for the selection of a nucleation event and nocturnal chemistry) that arise, mainly related to timing:

- What is the process producing the initial clusters? At what environmental conditions temperature, RH, radiation?
- Where (locally) does cluster formation happen and, in case, H_2SO_4 is involved, where is its origin?
- Are other compounds required, ammonia, water etc..?
- Why is cluster formation happening at this time and the corresponding chemical reactions are terminated after a few hours while precursors are still present?
- The model is running at least for one version with constant vapor concentration. Is this real?
- When H_2SO_4 is crucial for the cluster formation, why are there no more 5 nm particles appearing 12 hours after the peak of H_2SO_4?
- Why is the assumed H_2SO_4 concentration as high as measured peak values? Within the hours when nucleation mode particles grow in the model from 1 to 3 nm the H_2SO_4 concentration is only a smaller fraction of the model value. H_2SO_4 reaches model values at a time when already 10 nm particles are detected.

Response: As clarified above, the concerns of the reviewer on the aerosol dynamic models and their results are mainly based on misunderstandings of this manuscript. We have revised the manuscript to avoid such confusion.

There is no comparison between the aerosol size distributions and growth rates given by the aerosol dynamic models and obtained from the field campaign in urban Beijing. The models are used to test the formula and they are not based on the formula. Besides, the models are not used to investigate nucleation and growth mechanisms. A continuously increasing

growth rate was observed in this study, which is in accordance with previous literature. It took ~2 h for a 1.5 nm particle to grow up to 5 nm during the observed new particle formation event on Feb. 24, 2018, and this duration of 2 h was estimated from the measured aerosol size distribution rather than predicted by a model.

3D transport:

Looking into the experimental data given in Fig. 7 transport of externally produced particles and precursors is more likely to explain the observations (Cai et al 2018). Transport at night from elevated sources (~300 m) normally happens in clear air (low condensation sink) above the polluted nocturnal surface layer (Fig. 2). Vertical convection mixes rapidly (< 30 min) the air of residual and surface layers in the morning (Platis et al, 2016; Junkermann and Hacker, 2018). Contrary to atmospheric processes industrial nanoparticle sources run 24 hours, seven days a week and include sulphur and nitrogen chemistry as well as ammonia in large amounts (Bai et al, 1992).

A simple budget calculation with a box model similar like in Cai et al (2018) for a 300 km plume originating at Suizhong and Qinhuangdao spreading to ~40 km width (Rosenfeld et al, 2000), assuming a few hundred m residual layer (Platis et al, 2016, Junkermann and Hacker, 2018) and a replacement of the plume volume above Beijing with a wind speed of 25 km h^{-1} results, even without additional new particles, in number concentrations already in the order of magnitude finally observed in Beijing (> 80000 cm^{-3}) and in H_2SO_4 concentrations ~10^8 cm^{-3}, see also Junkermann and Hacker (2018). It is also in agreement with the diurnal pattern (Fig. 6) with convection beginning at ~08:00 local time and downwards mixing of a 200 m layer of this air mass into the low nucleation mode particle concentration 600 m surface layer (HYSPLIT) (Fig. 3). Numbers are based on measured and published emission data for power stations and from HYSPLIT meteorological data along the trajectory and allow at least a rough estimate of the magnitude of concentrations. How far new particle formation plays a role during the dry, cold and dark conditions during the transport and how aerosol size distributions and H_2SO_4 concentrations change would be a perfect task for a complete aerosol-chemistry-transport model. Such a plume production (Mohnen and Lodge, 1969) would widen the nanoparticle size distribution and provide additional particles in the lowest size bins. Field data show, that nighttime transport in the residual layer does not really lead to a massive loss of particles (Fig. 4). The plume hypothesis above is also in agreement with the results of spatial measurements of nucleation mode particles in Germany (Ma and Birmili, 2015) at stations surrounded by several fossil fuel burning power stations using SCR or SNCR technology.

Response: We agree with the reviewer that 3D transport may influence the measured aerosol size distribution and gas concentrations, especially for large (e.g., >5 nm) particles and trace gases with relatively long atmospheric residence time (e.g., SO_2). However, considering the atmospheric residence time of H_2SO_4 and sub-1.5 nm aerosols, the measured data shown in Fig. 7a in this manuscript is most likely a new particle formation event rather than a plume event. Deng et al. (2020b) reported that in urban Beijing, the atmospheric residence time of sub-1.5 nm particles are usually less than 5 min because of the relatively high coagulation sink. Note that there is no coal-fired power plant or cement plant inside the fifth ring road of Beijing. As a result, the observed H_2SO_4 and sub-1.5 nm particles were most likely to be formed near the station via daytime photochemical reactions rather than be transported from kilometers away.

In a perspective from long-term measurements in urban Beijing, the concentration of SO_2 in urban Beijing gradually decreased since 2012 due to strict emission control in northern China. An air pollution prevention and control action plan was conducted since 2013 and it led to a reduction of the $PM_{2.5}$ mass concentration in north China plain by more than 25%. However, the reduction of SO_2 and fine particle emissions and concentrations did not significantly change the frequency of new particle formation events in urban Beijing (Li et al., 2020). Hence, it is unlikely that the new particle formation measured in urban Beijing was often affected by industrial emissions.

In addition to confirming the local formation of new particles using aerosol size spectrometers down to the cluster size, the temporal evolution of the measured size distribution is a key to distinguish new particle formation events from plume events and traffic emissions. The examples given in Iida et al. (2008) show the identification of new particle

formation events in the polluted environment, and similar criterions were also summarized in Kulmala et al. (2012) and Kerminen et al. (2018). Figure R2 shows a plume event measured in urban Beijing. At 10:00, new particles were observed at ~3 nm and these particles grew steadily to ~20 nm at 12:00, which followed the typical pattern of a new particle formation event. However, starting from 14:30, new particles ranging from ~1 nm to ~10 nm with a higher concentration than the existing new particles were observed. Since ~10 nm particles were observed almost simultaneously with ~1 nm particles at 14:30 and there was no clear growth pattern in the measured aerosol size distribution, this event during 14:30 – 17:30 was likely to be a plume event. Note that since new particle down to ~1 nm were observed during this plume event, the source of this plume should be close to rather than away from the sampling site. In contrast, the formation and growth pattern in Fig. 7a indicates that the event observed on Feb. 24, 2018 was a regional new particle formation event.

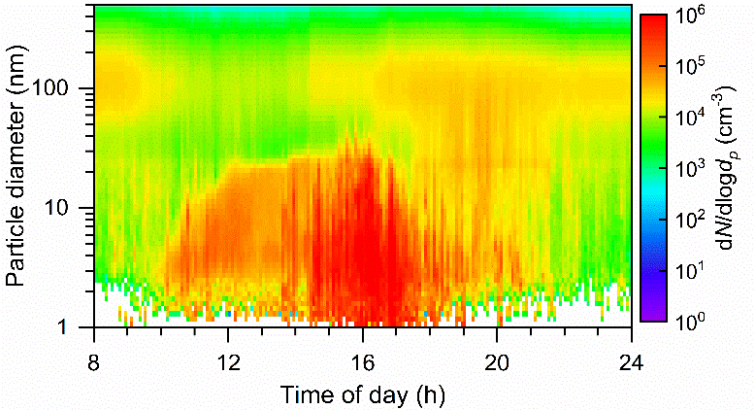

Figure R2. Aerosol size distributions measured in urban Beijing on Apr. 2, 2014.

Minor comments

Who is Julia (Page 7, line 1)

Response: We revised "Julia" as "the Julia programming language". Julia is the name of this programming language.

A reference to a paper under review (Cai et al, 2020) still has to be considered as grey literature as long as the review process is not open access. Also, as it is not clear whether the companion manuscript passes the review process, the information in this other paper is crucial for the current manuscript but might not be available. It could be placed in a supplement.

Response: We replaced Cai et al. (2020) with Deng et al. (2020b). Deng et al. (2020b) has been published and the details on the measurements are detailed therein.

A reference to a paper in preparation (Li and Cai, 2020) is not even grey literature!

Response: Li and Cai (2020) has been published now. The citation information is updated.

Define the conventional method, when it is first cited, Page 5, Lethipalo, 2014? Later in the paper it is confusing, with which model the current results are compared without a reference.

Response: Thanks. We added add a brief description of the conventional appearance time method in Lethipalo et al. (2014).

To clarify the used of the models, we added a paragraph at the beginning of Section 3.The introduction to the models in section 3.1 was revised as "A discrete aerosol model and a discrete-sectional aerosol model based on aerosol dynamics were used to provide an evolving aerosol size distribution and hence to test the conventional and corrected appearance time methods." Li and Cai (2020) is the reference for the discrete-sectional model, which also illustrates the principles of a discrete model. Note that these models were not compared to experiments, because they are used to generate an evolving size distribution rather than to investigate the nucleation and growth mechanism.

The same holds for the theory when the model, which is theory as well, is compared to another theory.

Response: The mechanism for the evolution of aerosol size distribution given by the model was not compared to other nucleation mechanisms. These aerosol dynamics models are used to provide growth rate in consistency with the simulated aerosol size distributions. Since the nucleation and growth mechanism used in these models do not affect this consistency, they do not affect the validity of these models in testing the formulae of the appearance time method. In Fig. 7b, the growth rate retrieved from the measured aerosol size distributions using the appearance time method is compared to the condensational growth rate contributed by H_2SO_4.

Within the time has to be defined in the text and figures. Is this Beijing CST?

Response: Yes. We revised the x-label in Fig. 7a as "Local time (HH:MM)". The time in Figs. 1 and 6 are the simulation time and it does not correspond to the time in a certain day. Hence, we keep the x-labels in Figs. 1 and 6.

Numbering of the tables. Why is the third table named table A1 instead of Table 3?

Response: We intend to put table A1 in the appendix. "A" is short for "appendix".

References

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