Response to Reviewer #1：

General comments:

The MS reports, analyses and discusses data on the NPF occurrence frequency, particle formation rate, growth rate, starting time, contribution of NPF events to UF particle number concentration, to CCN concentration and to aerosol extinction coefficient obtained within the German Ultrafine Aerosol Network for 5 years. The topic of the MS is timely and of interest for the international scientific community. The experimental and evaluation methods were carefully deployed and realised. The obtained results are very valuable considering both the spatial and temporal scales. They are also put into an international context by comparing them with those from some other, mainly central European sites. Nevertheless, there are several limitations which should be corrected and some aspects in which the MS could be and should be extended.

Response:

We appreciated reviewer's positive feedback and constructive suggestions which are of great value for improving the quality of our paper. Our point-to-point replies to the reviewer's comments are listed below.

Major concerns

1. The formation rate and growth rate were calculated for the diameter of 10 nm (Eqs. 1 and 2). Nevertheless, they are denoted as Jnuc and GRnuc. This is misleading since the subscript nuc usually indicates the properties at the nucleation, i.e. at a diameter of ca. 1.5 nm. Since the dynamic properties can strongly change with particle diameter in this range, this practice is not acceptable. The authors should use J10 and GR10 instead of the present notation all over the MS. More importantly and a consequence of this, it should be considered and discussed in more detail that the comparisons of these data to the other European results were accomplished at different diameters, e.g. to 3 or 6 nm. It is also noted in this respect that 1) the mean diameter (L144) should be replaced by the modal diameter, and 2) it should be specified for which particle diameter Dp was the CoagSDp calculated.

   Response:

   Thanks for the comments. We agree with the reviewer and have revised the text as below:

   (1) We have replaced J_{nuc} and GR_{nuc} with $J_{10\text{-}25}$ and $GR_{10\text{-}25}$, respectively all over the manuscript.

(2) In the description of calculation of growth and formation rates in Sect.2.3.2, the "mean diameter" has been replaced as "modal diameter" and the size of $CoagS_{Dp}$ was clarified as 10 nm.

(3) We have clarified the individual size range of GR and *J* in other European studies in Table S2 in the supplement material.

(4) We have extended the discussion on the comparison of GR and *J* between GUAN and other European sites in Sect.3.2. The revised paragraphs are as follows:

- Revision in Sect.2.3.2:

  "The growth and formation rate were evaluated for class I event in this study, while CS for all NPF events. The growth rate of nucleation mode particles ($GR_{10\text{-}25}$) is defined as the change rate of the modal diameter of the newly formed particles (Kulmala et al., 2012)…"

  "$$J_{10-25} = \frac{\mathrm{d}N_{10-25}}{\mathrm{d}t} + \mathrm{CoagS}_{10nm} \times N_{10-25} + \frac{GR_{10-25}}{\Delta D_{\mathrm{p}}} \times N_{10-25} \quad (2)$$

  where CoagS_{10nm} is the coagulation sink of particles with diameter of 10 nm, which can be calculated using the method proposed by Kerminen et al. (2001):

  $$\mathrm{CoagS}_{10nm} = \sum_{D_{\mathrm{p}}'=10nm}^{D_{\mathrm{p}}'=800nm} K(10nm, D_{\mathrm{p}}')\, N_{D_{\mathrm{p}}'} \quad (3)$$

  where $K(10nm, D_{\mathrm{p}}')$ is the coagulation coefficient between particles with sizes of 10 nm and D_{p}', and $N_{D_{\mathrm{p}}'}$ is the particle number concentration of particle with size D_{p}'."

- Extended discussions in Sect.3.2:

  "Figure 5 displays the annual GR measured at GUAN sites and other European sites (Boulon et al., 2011; Bousiotis et al., 2019; Bousiotis et al., 2021b; Herrmann et al., 2015; Kalkavouras et al., 2020; Lee et al., 2020; Manninen et al., 2010; Nieminen et al., 2014; Nieminen et al., 2018; Salma et al., 2016; Tröstl et al., 2015; Vaananen et al., 2013; Vana et al., 2016). For the GR values and the corresponding size range reported in those studies please refer to Table S2 in the supplementary material. The $GR_{10\text{-}25}$ for GUAN sites falls within the range of those reported in previous European studies. Caution should be taken that the differences in observation periods and size ranges of GR may influence the comparison among sites. In UB sites, the highest GR was reported at BUD, with the size range of 6–50 nm. LWE, KST and LTR showed the similar GR level, but the size range of GR at KST was 16.6–50 nm. The lowest GR in UB sites were observed at COP and HEL, with the evaluated size range of 5.8–30 and 3.4–30 nm, respectively. In RB sites, the GR at site CBW was about 6.6 nm h^{-1}, which was much higher than other RB sites. This high GR at CBW may be

resulted from the short observation period in this study, from 1 Apr 2008 to 31 Mar 2009 (Manninen et al., 2010). Meanwhile, another study reported the seasonal variation of GR at CBW between 10 and 25 nm as well (Nieminnen et al., 2018). The seasonal $GR_{10\text{-}25}$ ranged from 2.9 to 4.9 nm h^{-1}, which was similar with $GR_{10\text{-}25}$ at RB sites of GUAN. For LMT sites, the $GR_{10\text{-}25}$ at SCH and HPB were lower than the $GR_{7\text{-}20}$ at another two LMT sites PUY and OPM located in central France. Nieminen et al. (2018) also found that $GR_{10\text{-}25}$ at PUY was significantly higher than those at other LMT sites, possibly related to the vertical transport of particles within the boundary layer. For high altitude and remote sites, the $GR_{10\text{-}25}$ of ZSF was comparable to those of other sites."

2. The authors mention (L290) that the starting times (t1) of the NPF event were determined at different particle sizes of 5, 10 or 20 nm (lower diameter limit of the measurement setups), which makes the discussion and comparison difficult. This particle diameter variability could be taken into consideration in the first approximation when the starting times are shifted to the critical nucleation diameter of ca. 2 nm using the GR values which are the closest to the actual particle diameter by a subtraction of e.g. t1-(10-2)/GR10 (for 10 nm). This modified starting time tnuc could be compared more advantageously despite our knowledge on the GR in the diameter range from 2 to 3 or 5 nm is still inconclusive.

Response:

Thanks for the suggestion. We have converted the starting time from 10 nm ($t_{10\ nm}$) to 2 nm ($t_{2\ nm}$) according to the reviewer's suggestion. The hypothesis behind this estimation is that the $GR_{10\text{-}25}$ equals to $GR_{2\text{-}10}$. To verify the effectiveness of this conversion, we have compared $GR_{10\text{-}25}$ and $GR_{5\text{-}25}$ for MEL in the year of 2012 since the PNSD have been measured down to 5 nm. The relative difference between $GR_{5\text{-}25}$ and $GR_{10\text{-}25}$ is only 1 %, meaning only negligible bias will be introduced in the conversion.

The scatter plot of starting time $t_{2\ nm}$ is shown in Fig.R1. The $t_{2\ nm}$ was about 2 hours earlier than $t_{10\ nm}$. We have updated Fig.8 and corresponding text in Sect.3.3:

"Figure 8 shows the estimated starting time of class I events as a function of day of year. The starting time was initially estimated based on local time (UTC+1) and further converted to solar time according to the longitude of the sites. The PNSD observations in our dataset initiate from particle sizes from 5 or 10 nm at different sites (Table 1). However, starting time at 10 nm ($t_{10\ nm}$) was not able to describe the actual occurrence time of nucleation. Therefore, $t_{10\ nm}$ has been converted to the critical nucleation diameter of 2 nm ($t_{2\ nm}$) using the $GR_{10\text{-}25}$ values by $t_{2\ nm} = t_{10\ nm} - \frac{(10-2)}{GR_{10-25}}$.

Typically, most NPF events started between 07:30 and 9:00 solar time at all GUAN sites. Seasonal variations in starting time were evident, with earlier starting time in summer due to earlier sunrise. It is noteworthy that the differences in the starting time of NPF events exist between sites, as shown in Fig.8 and Fig.S7 in the supplementary material. The three mountainous sites (HPB, SCH, and ZSF) had the latest starting time around 09:00. Two UB sites LTR and LWE had the earliest starting time around 07:30. Starting time at BOS and three RB sites (MEL, WAL, and NEU) is around 08:30. Since the use of solar time has already eliminated the bias of local time relative to site longitude, the difference of starting time among sites mainly stems from the different diurnal variation of precursor concentration and CS, especially in urban background area."

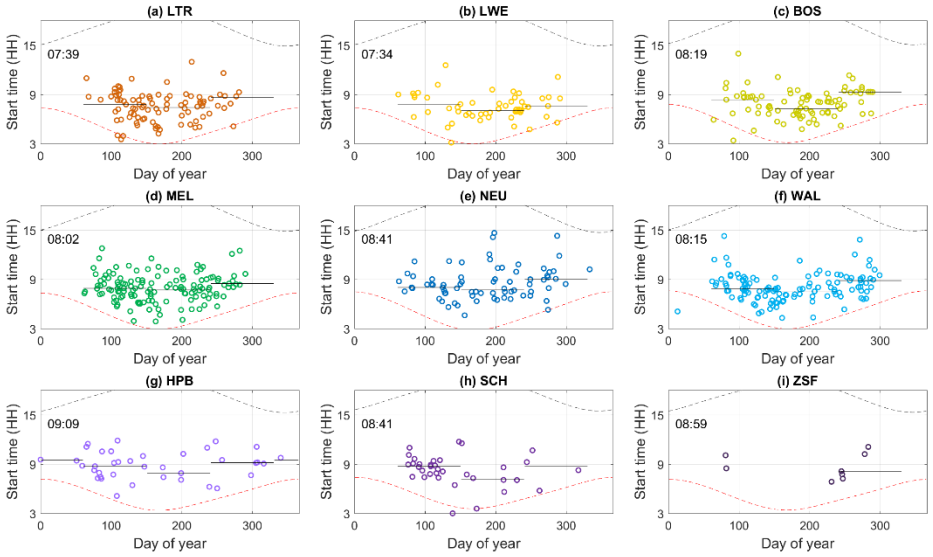

Figure R1: Scatter plot of NPF starting time at 2 nm depending on days of the year. The time displayed in the figure is the overall mean starting time. Black solid lines denote the mean value of starting time at each season, and the red and black dash line indicate the sunrise and sunset time, respectively.

3. Local time was used in the work as the time base (L136, L288). This selection (as all the other options as well) has both advantages and limitations. It should be clarified weather the starting times in Fig.8 were adjusted to clock change (expressed in UTC+1) or not. In relation to this, the reader may also wonder what the reasons were for selecting the 14-day smoothing in Fig.8 and not something else.

Response:

To better illustrate the influence of local environment on the NPF characteristics, the starting time evaluated in this study was the solar time, which was adjusted from local time

(UTC+1) according to the longitude of the measurement sites. We have emphasized this point in the text of Sect.3.3. Figure 8 has been revised accordingly.

"Figure 8 shows the estimated starting time of class I events as a function of day of year. The starting time was initially estimated based on local time (UTC+1) and further converted to solar time according to the longitude of the sites...

…Since the use of solar time has already eliminated the bias of local time relative to site longitude, the difference of starting time among sites mainly stems from the different diurnal variation of precursor concentration and CS…."

The reason for selecting the 14-day smoothing in Fig.8 is to show the seasonal variation of starting time. To make it clearer, we have replaced the 14-days smoothing to seasonal mean starting time, illustrated as black solid line in Fig.R1.

4. The authors are requested to explain why they can show occurrence frequency data for the winter months (e.g. Fig.3), while the column bars of nuc and GRnuc for winter are mostly missing in Figs. 6 and 7. Cf. lines 212-213.

Response:

Thanks for the comment. The GR and *J* were only estimated for class I NPF events. As shown in Fig.3, NPF events were observed in winter at sites NEU, WAL, HPB, and SCH. But the winter-time NPF events at NEU were all class II events, thus no winter-time growth and formation rates were estimated for NEU in Fig.6 and 7.

Accordingly, we have clarified this point in "Sect.2.3.2" and "Sect.3.2":

- Revision in Sect.2.3.2:

"The growth rate and formation were evaluated for class I event in this study, while CS for all NPF events."

- Revision in Sect.3.2:

"Figure 4 shows the basic statistics of annual $GR_{10\text{-}25}$, $J_{10\text{-}25}$ and CS at the nine GUAN sites. The growth and formation rate were only evaluated for class I event in this study, and the CS was estimated for all NPF event days…"

"Figures 6 and 7 present the seasonal $GR_{10\text{-}25}$ and $J_{10\text{-}25}$ at GUAN sites in this study. Since $GR_{10\text{-}25}$ and $J_{10\text{-}25}$ were only evaluated for class I events, there were NPF events observed in winter-time but no $GR_{10\text{-}25}$ and $J_{10\text{-}25}$ evaluated at some sites, for example at NEU."

5. The statement in L298-L299 is only partially acceptable. The occurrence and timing of the NPF events depend more sensitively on the ratio of the sources and sinks than on the

sources (precursor emissions) alone. The authors may want to add new aspects on the effects of the higher CS in cities.

Response:

Thanks for the suggestion. Following your advice, we have plotted the diurnal cycle of condensation sink (CS) on NPF days for all the nine GUAN sites in Fig.R2. Since the use of solar time has already eliminated the bias of local time relative to site longitude, the difference of starting time among sites mainly stems from the different diurnal variation of precursor concentration and CS. Unfortunately, we do not have measurements of precursors at the sites. It can be seen in Fig.R2 that the CS at UB sites increases rapidly during morning rush hour due to the strong traffic emission in urban area, implying also an increase of precursor concentration. The ratio of sources and sinks may be changed during this time period and further leads to earlier NPF starting time in urban area than RB sites. In the mountain area, the CS starts to increase at about 08:00 and reaches its daily maximum in the late afternoon, meaning that it may take some time for the development of the boundary layer and transport of the precursors upward after sunrise, resulting in late NPF starting time.

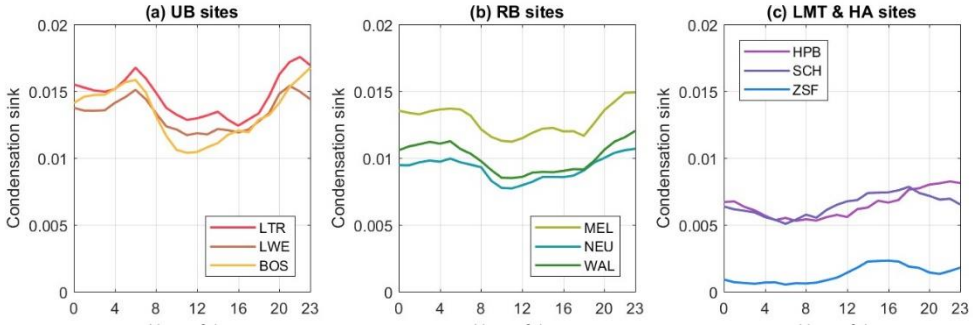

Figure R2: Mean diurnal cycle of condensation sink during NPF days at GUAN sites.

Accordingly, the text in "Sect.3.3 Starting time of NPF events" has been revised and Fig.R2 has been added as Fig.S8 in the supplementary material.

"…The three mountainous sites (HPB, SCH, and ZSF) had the latest starting time around 09:00. Two UB sites LTR and LWE had the earliest starting time around 07:30. Starting time at BOS and three RB sites (MEL, WAL, and NEU) is around 08:30. Since the use of solar time has already eliminated the bias of local time relative to site longitude, the difference of starting time among sites mainly stems from the different diurnal variation of precursor concentration and CS. Figure S8 shows the mean diurnal cycle of CS on NPF days for all the nine GUAN sites. CS at UB sites increases rapidly during morning rush hour due to the strong traffic emission in urban area, implying also an increase of precursor concentration. The ratio of sources and sinks may be changed during

this time period and further leads to earlier NPF starting time in urban area than RB sites. In the mountain area, the CS starts to increase at about 08:00 and reaches its daily maximum in the late afternoon, meaning that it may take some time for the development of the boundary layer and transport of the precursors upward after sunrise, resulting in late NPF starting time."

Minor comments

1. The Conclusion section is more a summary. It should be substantially shortened and the emphasis is to be put on the consequences of the actual results.

Response:

The conclusion has been re-organized, as follows:

"Based on a five-year dataset of the German Ultrafine Aerosol Network (GUAN), this study investigated the characteristics of NPF for various environments from urban background to high Alpine. The NPF occurrence frequencies show significant difference with respect to site categories, while the NPF occurrence frequencies at sites in the same category were found to be similar. Regional background sites had the highest NPF occurrence frequency, with an average value of about 19 %, followed by urban background sites with an average of 15 %. NPF events were observed on 7 % of days at low mountain range sites and only 3 % of days at the high Alpine site ZSF. The NPF occurrence frequencies at GUAN sites in this work were found to be in the range of the occurrence frequency at other sites in Central Europe reported in previous studies.

The annual mean growth rate for particle sizes of 10−25 nm ($GR_{10\text{-}25}$) varied from 3.7 to 4.7 nm h^{-1}, with minor differences among sites. The annual formation rate $J_{10\text{-}25}$ ranged from 0.4 to 2.9 cm^{-3} s^{-1}, increased with higher degree of anthropogenic emissions, implying the crucial role of anthropogenic precursors to NPF. The $GR_{10\text{-}25}$ for GUAN site falls within the range of those reported in previous European studies. Obvious seasonal patterns of $GR_{10\text{-}25}$ and $J_{10\text{-}25}$ were observed, with the highest in summer, and the lowest in winter for urban and regional background sites. Different seasonal patterns for the three mountain sites were observed, with the maximum in $J_{10\text{-}25}$ being reached in spring. Most NPF events started between 07:30 and 9:00 in solar time. Earlier starting time was found in summer due to earlier sunrise. The three mountainous sites had the latest starting time around 09:00. The two UB sites LTR and LWE had the earliest starting time around 07:30, while BOS and three RB sites around 08:30. The difference of starting time among sites mainly stems from the different diurnal variations of precursor concentration and CS.

The impact of NPF on ultrafine particles (UFPs), cloud condensation nuclei (CCN) and radiative forcing were quantitively evaluated and discussed. Over the entire observation

periods, the contribution of NPF on UFP was about 13 %, 21 %, and 7 % for the urban background, regional background, and low mountain range sites. The enhancement of CCN number concentration on NPF days was found to be the highest and the most significant in mountain sites. Similarly, the enhancement of aerosol extinction coefficient at 550 nm ($\sigma_{ext,550\ nm}$) on NPF days was respectively 1.4, 1.8, 1.6, and 1.9 at site NEU, HPB, SCH, and ZSF, while no statistically significant contributions were observed for the other sites. These findings underscore the importance of considering the local environments of NPF when assessing its potential impact on regional climate in models. They also emphasize the usefulness of a long-term aerosol measurement network with multiple sites for understanding the variation of NPF features and their influencing factors over a regional scale."

2. The authors may want to revisit their rounding off strategy at many places in the text and tables (e.g. 2.89 cm–3 s–1 in L25, Table 2), since the anticipated precision of these values seem exaggerated.

   Response:

   Thanks for the comment. We have checked and revised all the anticipated precision of numbers in the tables and text.

3. **L31-L32**. It is unusual to state that the particles are formed from precursors. Those chemical compounds (usually with longer atmospheric residence times) which yield the active players (usually with shorter residence times) in reactions are ordinarily called precursors. Thus, SO2 is a precursor compound, whereas its gas-phase oxidation product of H2SO4 is the vapour that plays an active role in the nucleation process. Clarification is needed.

   Response:

   We agree with the reviewer and revised the text as:

   "Atmospheric new particle formation (NPF) is a process initiated with the sudden formation of new particles with diameters less than 3 nm in the atmosphere. Low volatile gas molecules oxidated from gas-phase precursors cluster together and form new aerosol particles. These nano particles may subsequently grow into larger sizes by condensation or coagulation (Kulmala et al., 2014)."

4. **L32-L33.** The sentence is misleading. Condensation does not increase the particle number concentrations, while coagulation decreases them. These processes do not lead to growing particle number concentrations. Reformulation is required.

Response:

Thanks for the comment. This sentences have been revised as: "Atmospheric new particle formation (NPF) is a process initiated with the sudden formation of new particles with diameters less than 3 nm in the atmosphere. Low volatile gas molecules oxidated from gas-phase precursors cluster together and form new aerosol particles. These nano particles may subsequently grow into larger sizes by condensation or coagulation (Kulmala et al., 2014). The newly formed aerosol particles have the potential to contribute greatly to the number concentration of ultrafine particles (UFPs, particles smaller than 100 nm) or even larger sub-micrometer particles (particles smaller than 1 μm) (Ma and Birmili, 2015)".

5. **L90 and L125.** Check the citing format requirements of the journal.

Response:

The format of all the references have been checked and revised according to the requirement of ACP.

6. **L114.** Replace "Aerosol PNSD was measured" by "Aerosol PNSDs were measured", and similarly: "PNSD were generally measured" by "PNSDs were generally measured" (L118).

Response:

We have revised the expression of singular and plural words in the whole manuscript.

7. L158. The NSF was actually introduced and improve in Salma at al., 2017 and not in the reference cited.

Response:

Thanks. This article has been added in the reference list:

Salma, I., Varga, V., and Németh, Z.: Quantification of an atmospheric nucleation and growth process as a single source of aerosol particles in a city, Atmos. Chem. Phys., 17, 15007-15017, doi: 10.5194/acp-17-15007-2017, 2017.

8. The authors may want to write in the section title of 3.1 NPF "occurrence" frequency, and of 3.2 Growth and formation rate"s".

Response:

Thanks, and the titles of Sect.3.1 and 3.2 have been revised following the reviewer's suggestion.

9. **L243-L245**, twice. The order of the words and grammar should be checked: "the frequency of NPF event and undefined event are 14.5 % and 5.4 % for JFJ, respectively". Consider: the frequencies of the NPF events and undefined events were 14.5 % and 5.4 %, respectively for JFJ.

Response:

Thanks, and the sentences have been revised as:

"As stated by Herrmann et al. (2015), the frequencies of NPF event and undefined event are 14.5 % and 5.4 %, respectively for JFJ. The corresponding values are 3.3 % and 15.2 %, respectively for ZSF."

10. **L261**. The correct formulation is (Kulmala et al., 2022a, 2023).

Response:

Thanks, and the sentence has been revised as:

"Previous studies also found that GR varies little among different sites and exhibits only very weak dependency on the low-volatility vapor concentration, particularly in a fixed site (Kulmala et al., 2022a, 2023)".

11. **L342-L345**. Remove the considerable repetition.

Response:

Thanks. The sentence has been revised as:

"To evaluate the potential contribution of NPF to CCN, the relative enhancement of CCN number concentration (N_{CCN} enhancement, denoted as E_{Nccn}), which is the ratio between N_{CCN} after and prior to the NPF event, has been estimated following the approach proposed by previous studies (Kalkavouras et al., 2019; Ren et al., 2021)."

12. Caption of Fig.8. Correct: "…the red dash and black dash line indicate…".

Response:

Thanks. The caption of Fig.8 has been revised as:

"Figure 8: Scatter plot of NPF starting time (solar time) depending on days of the year. Black solid lines denote the mean seasonal starting time, the red and black dash line indicate the sunrise and sunset time, respectively."