

***We fully acknowledge the comments and suggestions of reviewer 1 and hope to have improved the (revised) manuscript accordingly.***

Crumeyrolle and co-authors present a measurement report on a long-term dataset from Lille, a large city located in France. The authors performed an extensive analysis of the particle number size distribution and meteorological parameters, in order to explain the factors involved in the new particle formation events over Lille. Crumeyrolle et al., reported that the majority of the NFP observations occurred during spring and summer. It was found that  $T > 275\text{K}$ ,  $\text{RH} < 45\%$ , and high solar radiation favored the NPF appearance. Additionally, the authors stated that despite the relatively high CS, new particle formation is observed. This observation is in concordance with other studies in large cities.

I very much appreciate the analysis of this long-term dataset. The results are clearly presented and extensively discussed. The manuscript is a valuable contribution to the field. I would recommend it to be published on ACP after addressing the following comments.

### **Specific comments:**

*Line 70: what do you mean by promoting growth rate? Do you mean, by promoting an increase in the growth rate?*

Initially the sentence was :

“A recent study (Bousiotis et al., 2021) using large datasets (16 sites) over Europe (6 countries) highlighted that solar radiation intensity, temperature, and atmospheric pressure had a positive relationship with the occurrence of NPF events at the majority of sites (exceptions were found for the southern sites), either promoting particle formation or growth rate.”

Now it reads:

“A recent study (Bousiotis et al., 2021) using large datasets (16 sites) over Europe (6 countries) highlighted that solar radiation intensity, temperature, and atmospheric pressure had a positive relationship with the occurrence of NPF events at the majority of sites (exceptions were found for the southern sites), either promoting particle formation or increasing growth rate.”

*Line 99: please define GAW.*

GAW definition (Global Atmospheric Watch) has been added to the manuscript

*Line 128: Please rephrase “SMPS dry (using a Nafion) particle number size distributions were also used for CS and growth rate (GR)”, you can delete growth rate and only use GR, since growth rate was defined before.*

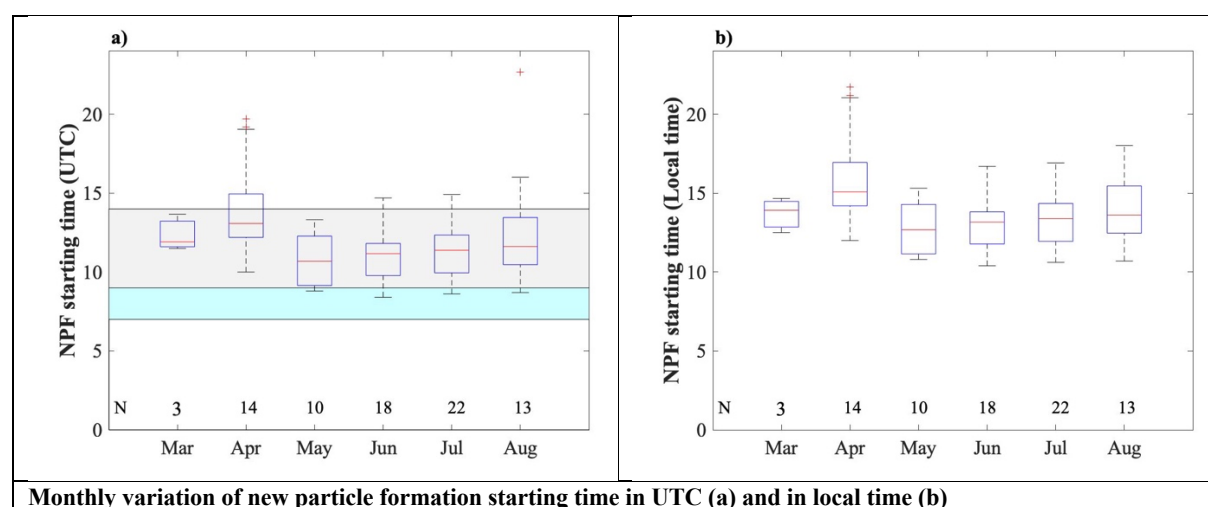
This was done.

*Line 192: can you please comment more about this “This might show that ATOLL is under the influence of air masses or particle and precursor sinks that favor the burst of UFP”. It is an interesting observation.*

The comparison between the study performed in Paris, 200 km south of Lille, and our results highlighted that bursts of UFP are observed more often over ATOLL than over Paris. As the paper focuses on NPF events and not burst, the authors have not yet looked in details at those burst events. These kinds of events may be linked to atmospheric dynamics as highlighted by Lampilahti et al. (2020 and 2021) and we plan to look into that since we purchased and installed in 2020 at the site an ultra-sonic anemometer. It could also be related to precursors concentration variations. However, at that time we did not have any instrument monitoring instrument. Since then, we also installed a SO<sub>2</sub> analyser. Therefore, in order to fully answer your comments, we would have to work on a more recent data set.

*Line 219: When the starting time and growth rate are discussed, the authors refer to the local time, not UTC time as in Fig. 3. This certainly is helpful to take into account the dynamic of the site where the monitoring was done. Can you please, refer to the caption of Fig. 4 that the time there is local?*

The time in the figure 4 was in fact UTC. Thanks for pointing that mistake in the caption. The reviewer is right to note that local time would make it easier to highlight the local dynamics. However, as most of the events (except two in March) are occurring during summer daylight saving time, the plot would be highly similar (see below) to the one presented in the submitted manuscript. To avoid any misunderstanding, the Figure 4 will remain in UTC time.



*Line 222: please define “colder period”. Is it colder than July? I think it is missing the comparative sentence. By looking at Fig. 4, to me, the “colder period” comprises March, April, and May. Or does it include winter?*

You are right, the colder period corresponds to March April and May. The results are also true for winter and fall but not shown here due to statistical issue (only few cases of NPF events). We added a sentence to clarify this point.

Previous sentence: “Indeed, the NPF starting time becomes earlier during the colder period and reaches a minimum in June (around 08:20).”

Corrected sentence: “Indeed, the NPF starting time occurs later during spring (also true for fall and winter) and reaches a minimum in June (around 08:20).”

*Line 231: here it is stated that the colder months are (March and November). I think the colder period should be more clearly defined otherwise it can be confusing.*

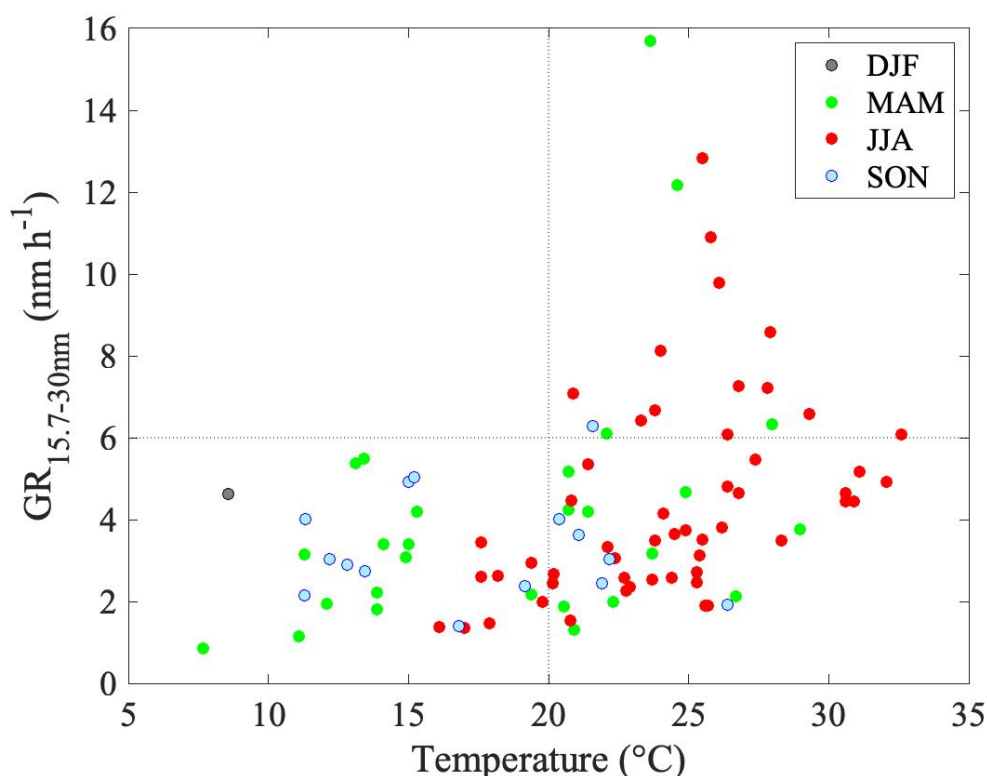
This sentence has been rephrased to avoid the used of colder months:” On average, NPF duration is shorter from May to August (around 8 hours) and increases up to around 13 hours on average in March and November.”

*Page 13: Can you please rephrase this sentence “GR<sub>15.7-30nm</sub> values were, in addition, plotted as a function of temperature for all years and seasons in Figure 5, which highlights that below 20°C, GR<sub>15.7-30nm</sub> values are lower than 6 nm.h<sup>-1</sup>, while, under warmer conditions (T >20 °C), GR<sub>15.7-30nm</sub> reach values up to 16 nm.h<sup>-1</sup>”.*

The sentence now reads :

“All GR<sub>15.7-30nm</sub> values were plotted in Figure 5 as a function of temperature. This figure highlights that GR<sub>15.7-30nm</sub> values are always lower than 6 nm.h<sup>-1</sup> for low temperatures (T<20°C), while, under warmer conditions (T >20 °C), GR<sub>15.7-30nm</sub> reach values up to 16 nm h<sup>-1</sup>.”

The figure was also modified to highlight the different seasons and highlight the tendency:



**New Figure 5 : Growth Rate ( $GR_{15.7-30nm}$ ) values as a function of ambient temperature for different seasons.**

*Page 13: can you please explain a bit more why you think this: “As previously observed in Figure 3a, the mean geometric diameters reached by the end of all NPF events are similar and averaged around 50 nm. This can be explained by the presence of a pre-existing mode of particles centered around 50 nm”. Thanks*

The second sentence could suggest that the growth of new particles was stopped at 50 nm because of the presence of pre-existing particles at this same size; this is not the message we wanted to give, so we removed the second sentence to avoid any misunderstanding. Explanations for the growth termination require a more detailed investigation in relation to the atmospheric dynamics (and in particular to the variability of the boundary layer height) and also in relation to the area where the process is initiated / takes place.

The authors plan in a future study to run Nanomap (Kristensson et al., 2014) on the data set to explore the area where the NPF events take place and to obtain statistics of the geographical occurrence of NPF events with dense spatial coverage and precision. NanoMap could also be used for the determination of the horizontal extent of an event combined with observations from site close to ATOLL such as Paris (200 km south) and Brussels (100 km east). This analysis could help us better understand regional events (Class I), in particular whether the growth stops at all sites simultaneously or only in Lille.

However in the absence of strong evidence supporting our assumptions, this sentence was reformulated to avoid any confusion: “As previously observed in Figure 3a, the median diameters reached by the end of all NPF events are similar and averaged around 50 nm. ”

*Caption Figure 6. The authors refer to spring (MAM, top) and summer (JJA, bottom) seasons. Is this classification related to the “warmer period”?*

We have checked throughout the manuscript for the occurrence of warmer/colder period wording in order to clarify this. In this case the warmer period (L.257) indeed corresponds to both spring and summer. “ This result is consistent with previous analysis performed over the boreal forest (Dada et al., 2017) and is linked to the fact that radiation seems essential for NPF during the warmer period (spring and summer), as the events occur almost solely during daylight hours (Kulmala et al., 2004).”

*Line 259: please rephrase this sentence “at moderately high RH ( $RH > 40\%$ ), hydrophilic aerosols could growth which will enlarge the sink for precursors and...”*

Initial sentence was: “at moderately high RH (RH >40%), hydrophilic aerosols could growth which will enlarge the sink for precursors and”

Now reworded as:” at moderately high RH (RH >40%), hydrophilic aerosols could grow which will enlarge the sink for precursors and”

*Line 260: can you please comment briefly on this? “high RH values limit some VOC (Volatile Organic Compounds) ozonolysis reactions, which further prevents the formation of condensable vapors necessary for nucleation”. How does the RH affect the ozonolysis reactions?*

More relevant references have been added. According to previous studies (Fick et al., 2003; Tillmann et al., 2010), the presence of RH can change the products of the ozonolysis. For example, Tillmann et al. (2010) have been running different ozonolysis experiments within the AIDA chamber at different temperatures and relative humidities. They found out that at high RH (68%), the  $\alpha$ -pinene is consumed and the pinonaldehyde is then formed. Since, this compound is mostly in the gas phase at room or higher temperatures, it won't generate Secondary Organic Aerosols (SOA) by condensation. Therefore, the RH can change the products formed via the ozonolysis reactions and tend to limit the production of SOA.

The sentence was changed to: “high RH values may limit the formation of some Volatile Organic Compounds (VOC) through ozonolysis reactions, inhibiting the formation of condensable vapors necessary for condensation.”

*Line 270: can you please mention that an example of biogenic compounds that inhibit NPF events is isoprene and cite Heinritzi et al., 2020 (Atmos. Chem. Phys., 20, 11809–11821, 2020)?*

This is now included : “As previously discussed, higher temperatures favor emission of biogenic precursors, including monoterpenes known to favor the occurrence of NPF event (Kulmala et al., 2004). Isoprene emission is also larger at higher temperature, but according to Heinritzi et al., (2020) its presence can make the difference between measurable new-particle formation events and their absence. Moreover, high temperature can also lead to evaporation of molecular clusters which may inhibit NPF events (Dada et al., 2017; Deng et al., 2020).”

*Can you please give more detail on how the CS was calculated? Please add this information to the method section. Thanks.*

We now included the equations used to calculate the CS :

“SMPS dry (using a Nafion) particle number size distributions were also used for CS ( $CS = 2\pi D \sum_i \beta_{Mi} d_{p,i} N_i$  Equation 1, where  $\beta_{Mi}$  is the transitional correction factor (Fuchs and Sutugin, 1970), the Knudsen number is  $Kn = 2\lambda_v/d_p$ , and  $\alpha$  is the accommodation coefficient and set to unity here)...”

$$CS = 2\pi D \sum_i \beta_{Mi} d_{p,i} N_i$$

**Equation 1**

$$\beta_{Mi} = \frac{1+K_n}{1+0.337Kn+\frac{4}{3}\alpha^{-1}Kn+\frac{4}{3}\alpha^{-1}Kn^2}$$

**Equation 2**

*In section 3.5, probably <https://doi.org/10.1038/s41586-020-2270-4> and Environ. Sci.: Atmos., 2022, 2, 491, can be useful for the discussion.*

Thanks for the interesting references that are now added to the manuscript. “Recent studies (Marten et al., 2022; Wang et al., 2020), performed in the CLOUD chamber, demonstrate that the presence of nitric acid (HNO<sub>3</sub>) and ammonia (NH<sub>3</sub>), typical within urban environments, contribute to freshly formed particles survival by increasing their growth rate.”

*The author may suggest that monoterpene emissions probably play a role in the observations. Since ATOLL is located at a peri-urban site, are there other precursors possibilities?*

Unfortunately, precursors were not measured over the period of interest here, thus the assumptions cannot be tested. The site is located inside a university campus with abundant tree plantings, which is why the monoterpenes were suggested as possible summertime precursors.

*For describing Fig. 3, the authors refer to the median, and later (on page 13) they refer to the mean geometric diameter. Can you please comment briefly on how those concepts compare? Are they similar or is there any conversion in between?*

When statistically describing (fitting) lognormal distributions, the geometric mean diameter of normal distributions is replaced by the count median diameter (CMD). In lognormal distributions, the log of the particle size distribution is symmetrical, so the mean and the median of the lognormal distribution are equal.

The median of the lognormal distribution and normal distribution are equal, since the order of the values does not change when converting to a lognormal distribution. Therefore, for a lognormal distribution,  $D_g = \text{CMD}$ .

$$D_g = \text{CMD} = (D_1^{n_1} \cdot D_2^{n_2} \cdot D_3^{n_3} \dots D_N^{n_N})^{1/N}$$

where:

$D_g$  = geometric mean diameter

$D_i$  = midpoint particle size

$n_i$  = number of particles in group  $i$  having a midpoint size  $D_i$

$N = \sum n_i$ , the total number of particles, summed over all intervals

In this case, we did not fit the aerosol SD but we used the median diameter, so we removed the term “mean geometric diameter” and replaced it by ‘median’.

**Technical comments:**

*Line 17: please add a dot after “particles” to finalize the sentence.*

Done

*General: please define how the abbreviations are written, condensation sink (CS) or Condensation Sink (CS)? the same applied to other abbreviations such as UFP, GR, etc.*

Done

*Line 23: probably it is better to write: using a 4-year long-term dataset, without mentioning the exact date (in the abstract).*

Done

*Line 29: please change Growth Rate to growth rate.*

As we choose to define the abbreviations using capitals, we kept the Growth Rate as is.

*Line 32: please change “reaches” to “reached” to keep the abstract in past.*

Done

*Line 35: please change “New Particle Formation” to “New particle formation” to be consistent with the abstract.*

Again, As we choose to define the abbreviations using capitals, we kept the New Particle Formation as is and only use NPF in the following sections.

*Line 36: Please change “The latter” to for example “These particles”.*

Done

*Line 39: Please rephrase this sentence “The freshly-formed particles then grow to larger sizes, from a few nm in particle diameter up to sizes ( $D_p > 100$  nm) at which they may act as cloud condensation nuclei (CCN,..”. A possibility could be “The newly-formed particles then grow to larger sizes ( $D_p > 100$  nm) at which they may act as a cloud condensation nuclei (CCN).*

It was corrected as suggested.

*Line 52: There is a parenthesis missing at the end of the sentence “Differences were found in both the seasonality and intensity of NPF events according to the site type (urban, traffic, regional, background, rural, polar, high altitude (Dall’Osto et al., 2018; Sellegri et al., 2019)”.*



This was added.

*Line 124: can you please add which diameter range you consider to be Aitken mode?*

The new sentence now reads: ‘and their consequent growth to Aitken mode ( $D_p < 80$  nm)’

*Line 67: please change “A recent study (Bousiotis et al., 2021) using large datasets (16 sites) over Europe (6 countries) highlighted...” to “A recent study (Bousiotis et al., 2021) used a large dataset (16 sites) over Europe (6 countries) and highlighted that...”*

Corrected as suggested.

*Line 95: please change “(Métropole Européenne de Lille, more than 1.1 million inhabitants)” to “(Métropole Européenne de Lille with more than 1.1 million inhabitants)”*

Corrected as suggested.

*Line 112: please rephrase “The instruments use in this study focused on aerosol properties including number size distributions, chemical composition, and optical properties, and details are described below”. For example, “The instruments used in this study measure the aerosol properties including number, size distributions, chemical composition, and optical properties. The details are described below”.*

It was corrected as suggested.

*Line 119: please rephrase “Typically, the scan time was chosen to be 300 seconds. To take into account the multiple charge effect and the losses through diffusion, particle concentrations were corrected using the equation given by the manufacturer specifications (AIM 10.2.0.11)” to e.g.,*

*“The scan time was 300 seconds and the particle concentrations were corrected by taking into account charge effects and diffusion losses”.*

This was modified into : “The scan time was 300 seconds and the particle concentrations were corrected by taking into account charge effects and diffusion losses calculated using the manufacturer software and algorithms (AIM 10.2.0.11).”

*Line 129: please change “which” to “which”.*

Done.

*Line 134: please rephrase “First, the NPF starting time was identified when the newly formed mode was observable in the first bins of the SMPS (15.7 nm) and the time of peak concentrations for particles with a diameter of 30 nm ( $N_{30}$ ) during NPF were manually identified” to “First, the NPF starting time was identified when the newly formed mode was observable in the first bins of the SMPS (15.7 nm). The final time was manually selected and it was defined as the time at which the particle concentration of 30 nm-particles reached a maximum”. (For example).*



It was corrected as suggested.

*Line 144: please change “5 L.min<sup>-1</sup>” to “5 L min<sup>-1</sup>”  
General: sometimes it is written X % and other times X%, please be consistent.*

We modified the flow units and checked the whole manuscript to keep percentage as xx %.

*Line 173: please change “(polar, rural, high altitude, remote, urban)” to “(polar, rural, high altitude, remote, and urban)”*

It was corrected as suggested.

*Line 205: please change “(See supplementary materials)” to “(see supplementary materials)”*

Done.

*General: There is inconsistency in the font size and font type used along the manuscript, please unify.*

We are so sorry for that and we of course modified it in the revised version of the manuscript

*Y label of Fig. 4b and along the manuscript: please change “(nm.h<sup>-1</sup>)” to “(nm h<sup>-1</sup>)”.*

It was corrected as suggested.

*Page 13: please change “Over urban areas (Beijing or Shangai)” to “Over urban areas such as Beijing or Shangai”.*

Done

*Figure 3. Do the plots shown here represent an average or are they representative examples?*

To plot Figure 3, we used all the data recorded during spring and summer. We selected all days when a NPF event was observed and then we averaged the data to one-hour time resolution using median filtering as in Kulmala et al (2022). This was clarified within the manuscript. “Median daily contour plots of the particle number size distributions (PNSD) obtained from the SMPS are shown in Figure 3 separately for NPF event, undefined and non-event days observed during the warm period (only spring and summer). All the aerosol size distributions observed during NPF event (around 800 PNSD), undefined (around 2300 PNSD) and non-event (around 1700 PNSD) days were selected then averaged to one-hour time resolution using median filtering.”

*Figure 5. please change “(nm/h)” to “(nm h<sup>-1</sup>)”.  
Figure 6b: please change “(W/m<sup>2</sup>)” to “(W m<sup>-2</sup>)”.*

The units were changed accordingly to the suggestions.

*Line 247: please change “total solar radiations” to “total solar radiation”.*

Done

*Line 255: there is one line spacing missing between studies and (Duplissy...). Line 262: please change temperature conditions (T) to (T).*

This was corrected.

*General: I would recommend increasing the font size of the axis on the plots, for example in Figure 6b, Figure 7, etc.*

We did increase the font size on all diel figures. We also added a grid to improve the readings.

*Line 276: please change “(larger than  $2 \cdot 10^{-2} \text{ s}^{-1}$ )” to “(larger than  $2 \cdot 10^{-2} \text{ s}^{-1}$ )” or “(larger than to  $2 \cdot 10^{-2} \text{ s}^{-1}$ )”. The same for lines 278 and 279.*

This was corrected in  $2 \times 10^{-2} \text{ s}^{-1}$

*Line 359: typically, there is a space between the number and the unit, e.g., 10 h instead of “10h”.*

The error was corrected throughout the manuscript.

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