We would like to thank the reviewer 2 for his comments and his/her suggestions that have improved the quality of the manuscript.
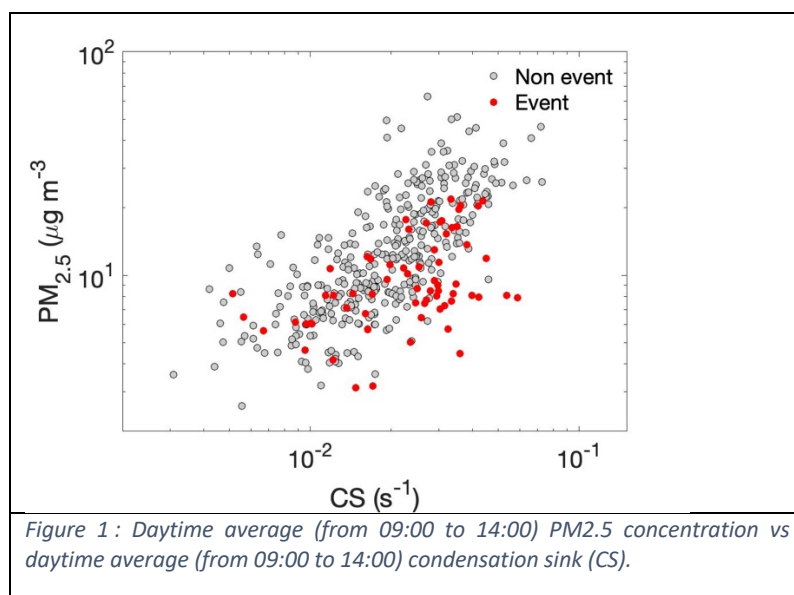
This work reports long-term (4 years) measurements of particle number size distribution at an urban site in Lille, North of France. This study aims to better understand the environmental factors favoring/disfavoring atmospheric new particle formation in this urban environment. These studies allow to reduce the lack of knowledge that still exist on the process of new particle formation and their subsequent growth. It is a complex and extended dataset and analysis and the results will fit within the scope of ACP, being of interest for the international research community. However, I would suggest some aspects to be considered in order to improve the manuscript and/or strengthen its impact before it is published in ACP.

Major comments

The dataset presented is of interest for the international community and combine a large number analysis. However, the manuscript is mainly descriptive, the results of each section are not analyzed/discussed in deep and not big conclusions are reached. I could suggest the authors to include more discussion about GR (maybe the contribution of H2SO4 to GRs and/or improve the discussion of possible precursors -comment below-), include Formation Rate analysis, the differences between event and non event days (in deep analysis) and try to investigate the differences on the CS. In this sense, since 1) the introduction is mainly focus on urban areas and CS effect on NPF events and 2) ATOLL has some measurements of aerosol chemical composition (with ACSM), I would also suggest to look if there is some relationship with the chemical composition of pre-existing particles acting as CS (two recent reads about CS efficiency Du et al. (2022) and Marten et al. (2022)). The ACSM measures from ~80nm and it's a good estimation of the CS chemical composition. This links with the fact that BCwb is high during non-event days, and recent study (Yus-Diez et al., 2022) have shown the impact of secondary aerosol on this quantity.

Thanks for this comment. At the moment the authors wrote this manuscript those papers were probably not yet published. However, their results are extremely interesting and need to be included into our manuscript.
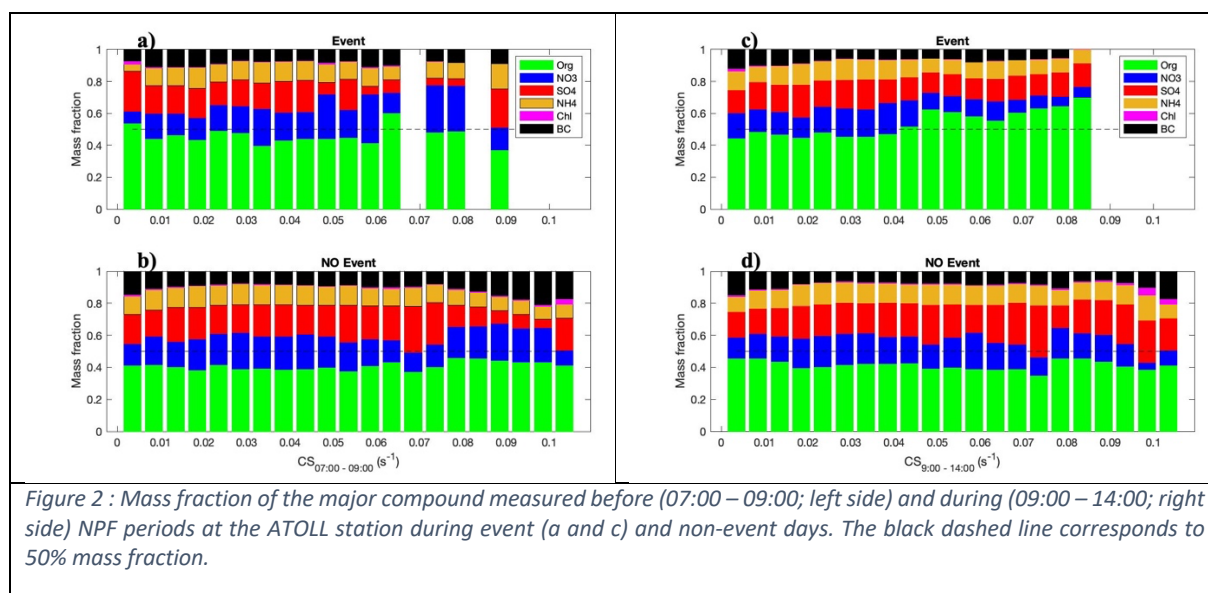
The Figure 1 is showing the $PM_{2.5}$ as a function of the CS both averaged over the period where most of the NPF are observed (09:00 to 14:00) for NPF event (red) and non-event (grey) days. This figure similar to the one on Du et al. (2022) results is clearly different from the Beijing situation. Indeed, over Beijing the NPF event are clearly associated with lower values of CS and low $PM_{2.5}$ concentrations. Over ATOLL, the NPF events are not clearly associated with low CS values. In fact, NPF events and non-events are occurring over the same range of CS values (0.03 – 0.7 s^{-1}). However, one can see that the $PM_{2.5}$ concentrations are on average lower during NPF event days in comparison to NPF non-event days.

Figure 1 : Daytime average (from 09:00 to 14:00) PM2.5 concentration vs daytime average (from 09:00 to 14:00) condensation sink (CS).

An ACSM is monitoring the aerosol chemical composition on the ATOLL station. Du et al (2022) highlighted the chemical composition of the particles that contribute to the CS. The comparison of the chemical mass fraction observed during NPF event days and non-event days highlight the most effective chemical mass fraction in taking up condensable vapors. They observed a large increase of nitrate and a decrease of organics with the CS values. Moreover, the chemical mass fraction highlights a large increase of Nitrate during non-event days in comparison to NPF event days. As nitrate is highly hygroscopic, nitrate enriched particles are more likely to adsorb water and grow to larger sizes promoting the uptake of gaseous vapors. These observations, consistent with simultaneous kappa calculations, suggest that the particle chemical composition affects the efficiency of the CS for both condensable and reactive uptake of vapor molecules.

In our study, we highlight that the CS is largely influenced by the freshly formed particles, so Figure 2 presents the chemical composition of the particle as a function of CS during two specific periods: before (07:00 – 09:00) and during (09:00 – 14:00) the NPF periods for NPF event and non-event days (Figure 2). During non-event days, both periods (07:00 – 09:00 and 09:00 – 14:00) exhibit similar mass fraction of all compounds with on average 41% Organics, 16% of nitrate, 21% of sulfate, 11% of ammonium, less than 1% of chloride and around 10% of Black Carbon. As the aerosol sources during non-event days are supposed to be the same throughout the day, this result was actually expected.

The same comparison, for event days, shows a larger contribution of Organics to the CS during the NPF period (54% on average) in comparison to the period right before the start of the NPF events (46% in average). Indeed, for large values of CS (> 0.045) the contribution of Organics is larger than 50%, reaching a maximum of 69% for CS of 0.085 s^{-1}. This result suggests that organic vapors are likely involved in the particle growth.

Figure 2 : Mass fraction of the major compound measured before (07:00 – 09:00; left side) and during (09:00 – 14:00; right side) NPF periods at the ATOLL station during event (a and c) and non-event days. The black dashed line corresponds to 50% mass fraction.

Part of this discussion has been added to the supplementary material as it contributes to providing clues on the involvement of organic compounds in particle growth. However, we believe that a deeper analysis would be required to fully explore the role of the particle chemical composition on CS efficiency, and, similar to the referred papers (Du et al. 2022, Marten et al. 2022), this could be the subject of a dedicated study. Moreover, the relevance of such a detailed study in the present work might be questionable since the CS does not appear as a limiting factor for the occurrence of NPF, which remains the main focus of the paper.

Minor comments
L36 – "highly // significantly" contribute.
This was corrected
L63 – I would use "NPF event" instead of "nucleation event", nucleation is just the process of formation.
Of course, the reviewer is right and this has been corrected as suggested.
L71 – I would include reference (Dada et al. 2007 fits well).
The reference was included to support the link between high temperatures and the growth of newly formed particles
L94 – Indicate altitude
The altitude of the station has been added.
L97 – This reference is not included in the reference list. Include at least title?
The reference has been added.
Velazquez-Garcia[1,2]*A., S. Crumeyrolle[2]*, J.F. de Brito[1], E. Tison[1], E. Bourrianne[2], I. Chiapello[2], V. Riffault[1]. Deriving composition-dependent aerosol absorption, scattering and extinction mass efficiencies from multi-annual high time resolution observations in Northern France. Submitted to Atmospheric Environment, Apr. 2022.

L99 – Rose et al. 2021 maybe fits better?
The authors choose to keep Laj et al. and add Rose et al.
L104 – volcanic plumes affect the surface levels?
Yes, the authors added a reference (Boichu et al. 2019) in which they explore month/year long ACSM data in France from stations in contrasting environments. They have shown that volcanic

sulfate aerosols exhibit a distinct chemical signature in urban/rural conditions, with NO_3:SO_4 mass concentration ratios lower than for non-volcanic background aerosols.

L115 – 3082 is not a DMA, is the classifier. Maybe, DMA would be 3081A?
You are right, the SMPS is a 3082 but the DMA is a 3081A. This sentence has been corrected.

L123 – reference format and rephrase.
The sentence is now : "The classification procedure, presented in Dal Maso et al. (2005), is following the decision criteria based on the presence of fine particles (Dp < 25 nm) and their consequent growth to Aitken mode (Dp < 80 nm)."

L129 – Which are the properties of the CS? The same properties than the vapour? The loss rate will depend on the molecular properties but also the CS chemical composition. Same line, "which" underlined.
Reviewer 1 suggested to include the equations used to describe the CS calculations. So we included both transitional correction factor (β) and CS equations into the revised version of the manuscript. The Knudsen number is defined as the ratio of the molecular mean free path length to a representative physical length scale. The mean free path here was then calculated assuming all condensable vapors were sulfuric acid. Therefore, we used the molar mass and the diffusion volume of sulfuric acid. For those calculations, the chemical composition of the preexisting particles is not considered. However, as the reviewer 2 suggested, we added a part to describe the CS chemical composition and it's influence on the condensation efficiency.

L131,132 – The CS only indicate the loss by condensation, no by coagulation.
Right, the sentence was corrected accordingly. We meant to say that the more preexisting particles, the larger the probability for clusters to coagulate on it.

L133, 137 – Please rephrase and indicates the meaning of GR15-30 (or don't use it and just indicate that the GR will always refer to the growth rate of 15-30nm).
The whole paragraph was modified into :
"The particle $GR_{15.7 - 30}$, from 15.7 to 30nm, was calculated based on the maximum-concentration method described in (Kulmala et al., 2012). First, the NPF starting time was identified when the newly formed mode was observable in the first bin of the SMPS (15.7 nm). Then, the time when the concentrations for particles with diameter of 30 nm (N_{30}) peaked was alsomanually identified. Particle $GR_{15.7 - 30}$ was then calculated by linear regression of particle size vs. time span from the NPF start until time when N_{30} reaches a maximum ($GR = (D_{p,2} - D_{p1})/(T_2 - T_1)$)"

The

The

"SMPS dry (using a Nafion) particle number size distributions were also used for CS ($CS = 2\pi D_i \beta_{Mi} d_{p,i} N_i$ Equation 1, where β_{Mi} is the transitional correction factor (Fuks and Sutugin, 1970), the Knudsen number is $Kn = 2\lambda_v / d_p$, and α is the accommodation coefficient and set to unity here) and GR calculations. CS estimates the loss rate of the condensable vapors (Kulmala et al., 2001) which were assumed to have molecular properties similar to sulfuric acid (Dal Maso et al., 2005). A high CS indicates the presence of large surface area of aerosol particles onto which NPF precursors can condensate. The particle $GR_{15.7 - 30}$, from 15.7 to 30nm, was calculated based on the maximum-concentration method described in (Kulmala et al., 2012). First, the NPF starting time was

concentration method described in (Kulmala et al., 2012). First, the NPF starting time was identified when the newly formed mode was observable in the first bin of the SMPS (15.7 nm). Then, the time when the concentrations for particles with diameter of 30 nm (N_{30}) peaked was alsomanually identified. Particle $GR_{15.7 - 30}$ was then calculated by linear regression of particle size vs. time span from the NPF start until time when N_{30} reaches a maximum ($GR = D_{p,2} - D_{p1}/T_2 - T_1$)

$$CS = 2\pi D \sum_i \beta_{Mi} d_{p,i} N_i \qquad\qquad\qquad \textit{Equation 1}$$

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

144,145 – "L.min" change by "L·min". "Wood Burning" not necessarily capital letters.
This was corrected

L148, 149 – indicate what is each instrument measuring and how you use it. About the solar radiation, only global seems to appear in the results sections.
The solar irradiance on the horizontal plane is measured every minute with two instruments, a pyrheliometer and a pyranometer. Both are associated with an EKO automated sun tracking system and both are Kipp&Zonen instruments (CHP1 and CMP22) with a close spectral range and response (around 300 nm to 3600 nm at half maximum). The pyrheliometer provides the direct normal irradiance (DNI) and the pyranometer measures the diffuse horizontal solar irradiance (DifHI) using a sun shading ball. The global horizontal solar irradiance (GHI) is obtained as follows: GHI = DNI x cos(SolarZenitalAngle) + DifHI. The quality of the instruments and the automated shading device ensures the accuracy of the measurements to around 1% for GHI under clear sun conditions, and several percents for GHI and DifHI depending on the magnitude of signal (and the presence of clouds).

The sky imager is a camera equipped with a fisheye lens to cover the entire upper half sphere. The cloud cover is estimated from an algorithm comparing the different values of the red, green and blue components of each pixel of the taken images.

Part of this discussion was added to the manuscript :
"Solar radiation at the surface are measured every minute at the sampling site using a set of Kipp & Zonen pyranometer (CM22, for diffuse fluxes using a sphere shadower) and Normal Incidence pyrheliometer (CH1 for direct fluxes), the solar radiation being then calculated as the sum of the diffuse and direct fluxes. A sky imager (Cloudcam, CMS) is a camera equipped with a fisheye lens to cover the entire upper half sphere. The cloud cover is estimated from an algorithm, named Findclouds and provided by the manufacturer, comparing the different values of the red, green and blue components of each pixel of the images taken (Shukla et al., 2016)."

L158 – section is titled "NPF event frequency and Growth rate" but the growth rate is not included in this section.
The title was modified into : "NPF event frequency"

L169, 172 – I would not compare the undefined frequency with the boreal forest, the environments are pretty different. Have the authors other explanation? Maybe the NPF doesn't growth enough (cut off Dp is 15.3nm)? Similarly for the whole text, the authors compare with

A large part of the published NPF studies are based on observations performed within the boreal forest, which is why we use those studies for comparison. However, we do understand the importance of comparing our results to similar site types. Throughout the manuscript, we also compare our results to NPF observation in urban environments such as Dos Santos et al. (2015) performed in Paris but also observations performed over Chinese megacities or other megacities in Europe (Beijing, Helsinki, London, Leipzig, Copenhagen, Granada, etc..). In particular the paragraph following this one is dedicated to the comparison of event, non-event and undefined frequencies to those observed in Paris.

The undefined events are defined by Dal Maso et al. as fine particles (Dp < 25nm) that does not grow during their existence. The reviewer is right to highlight the fact that these particles may have stopped growing below the minimum cutoff size of 15.7nm so with our measurements we could have missed the growth of those particles.

We corrected the 3.2 section title as suggested by the referee. We are following the ACTRIS standards by using a Nafion membrane up-stream the SMPS. We removed dry from the manuscript and added one sentence in the description of the SMPS:

"The Scanning Mobility Particle Sizer (SMPS) measures particle number size distribution between 15.7-800 nm downstream a Nafion membrane as recommended by ACTRIS standards to keep relative humidity below 40%. "

Reviewer 1 also commented on those figures and we have modified the figure caption and the text accordingly.

" Figure 3: Hourly median particle number size distribution (15.7 nm<Dp<800 nm) observed during NPF event (a), undefined (b) and non-event (c) days in spring and summer from 2017-2020."

Main text: "Median daily contour plots of the particle number size distributions (PNSD) obtained from the SMPS are shown in Figure 3 separately for NPF event (n ≈ 800 PNSD), undefined (n ≈ 2300 PNSD) and non-event (n ≈ 1700 PNSD) days observed during the warm period (only spring and summer). The PNSD were first selected then averaged to one-hour time resolution using median filtering."

This was corrected.

Right.

Reviewer 1 also noted this annoying switch from GR to GR15.7 -30. The authors carefully went through the whole manuscript to remove the occurrence of GR when it was referring to our results.

L231 – "presence of availability"?
This is a typo. We meant presence or availability. Thanks for pointing it out.

Figure 4 – the number of cases for the GR is the same than for the starting time figure?
Yes, the number of events per month is the same for all this study. Therefore, we did not reproduce the N on Figure 4b.

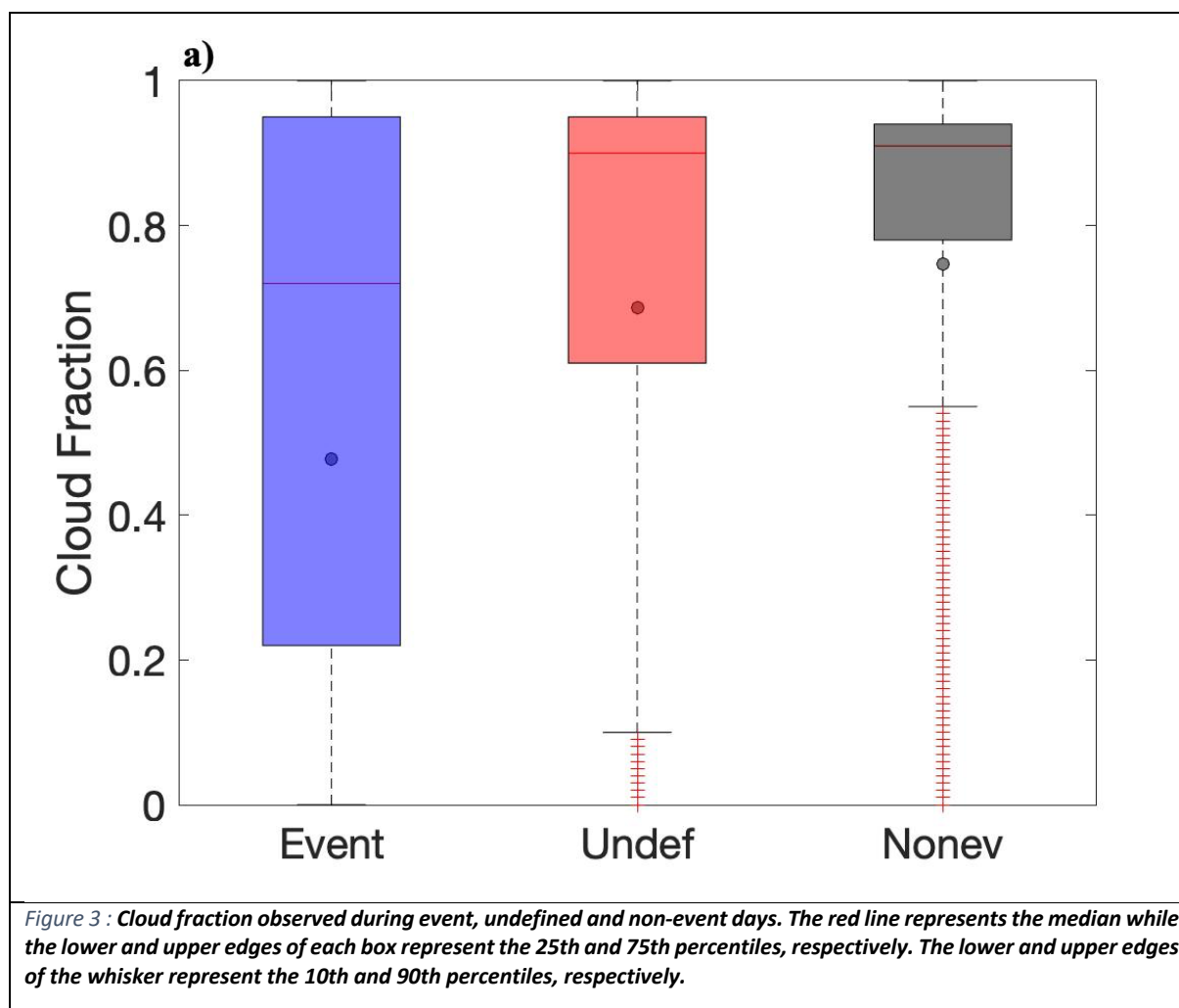Lines missing in page 13, but "nm.h-1" again change "." by "·". Temperature "dependence".
All these mistakes were corrected.

Finally, in this same page, the authors points to the importance of biogenic emissions, however, the measurements were done in an urban environment probably where higher influence of anthropogenic organic compounds are expected? As I pointed in my major comment, what is the contribution of H2SO4? Should be minimal I guess (even more at this size range).
The site is located on the university campus with abundant tree plantings, and about 4 km from the city center. By comparison of several parameters (SSA, scattering coefficient, number concentration etc.) observed over ATOLL to the recent reviews of Laj et al. and Rose et al. (2021), the site could indeed be considered as close to urban sites. However, the various tree varieties can emit large concentration of organics, especially in summertime. The results shown in Figure 2 of this document clearly highlight that organics have a clear role in the growth of freshly nucleated particles, and that sulfate-related particles are less important in case of NPF event days.

Figure 6 – I would recommend showing same periods for figures a) and b), if not the reader cannot use both figures information. In this sense, include two boxplot figures (one for each period) or combine solar radiation figures on only one. In addition, here the authors use UTC, and before have been using Local Time, please use always the same and indicate it (e.g. L238). Finally, figure caption is not really clear, I would add c) instead of "b) top and bottom".
All the times used in the manuscript and figures are in UTC, and this mistake was corrected in the revised version. We plotted the cloud fraction over the whole day (Figure 3).

Figure 3 : Cloud fraction observed during event, undefined and non-event days. The red line represents the median while the lower and upper edges of each box represent the 25th and 75th percentiles, respectively. The lower and upper edges of the whisker represent the 10th and 90th percentiles, respectively.

Results are similar but the authors truly believed that the most relevant plot is the one already in the manuscript. Indeed, the cloud fraction can change quite quickly during the day. In the morning or in the late afternoon it is not unusual to observe stratiform clouds in Lille. For our study, we know that the cloud cover will influence the NPF occurrence but mostly when the NPF events occur (from 09:00 - 14:00). This is why the authors would like to keep the figure as it is.

L255 – space before reference
Done.

L256, 261 – I don't see the link of these reasons with the data shown on Fig 7. RH<40%, these relative humidity values are not observed at your site. High RH limit some VOC ozonolysis, but what about H2SO4–water nucleation? The authors linked in previous section the GR with the increase of temperature, what about the other parameters?

Within the instrument the RH is always lower than 40% according to ACTRIS standards. However, the RH is also monitored via a meteorological station shown in Figure 7. One can clearly see on this figure, that the event days are associated with lower RH for both seasons in comparison to non-event days. This effect has been observed in many other studies (Bousiotis et al., 2021; Dai et al., 2022; Lv et al., 2018; Baalbaki et al., 2021) performed within real atmosphere. The reasons might be those cited within the manuscript.

Some laboratory measurements performed over a large range of RH values (Yu et al., 2017) observed that the growth rate of sub 3nm particles might be more important (1.5 nm h^{-1}) by a factor of almost 2 at high RH (80%) in comparison to low RH (10%). However, according to our result the growth of UFP (below 30nm) might be due to organic vapors.

Based on the results obtained in the CLOUD chamber, RH is expected to enhance the particle formation rate of the H2SO4-water binary mechanism (Duplissy et al., 2016). At ATOLL, we do not measure H_2SO_4 so we do not know how much H_2SO_4–water nucleation is involved in the first phase of the NPF events observed. However, according to the work of Dunne et al. (2016), we do not expect that this binary nucleation mechanism would play a central role at our site, since it is a priori not very efficient in the boundary layer.

Other parameters (NO2, ozone, Absorption and Scattering Angstrom Exponent, aerosol chemical composition, effective diameter, etc…) were investigated to better understand the GR variation as a function of temperature. Some expected tendencies were observed such as more BCwb when the temperature is low. However, we did not find any other parameter to better explain the GR variation with temperature. This might be related to the emission of Organic precursors that is not yet measured over ATOLL.

To answer to both comments (H_2SO_4–water nucleation and GR increase with temperature), precursor measurements are needed. The deployment of instruments allowing the identification of the nature of the species involved in the process is planned on the site, and will make it possible to answer these questions in the future.

Figure 7 – please use same scale for both periods. Again UTC.
As previously said all the time are in UTC. The scales have been modified.

L273, 279 – It is not clear what are you comparing. For event days are larger, maybe yes, but I can not see the overall differences, just the diurnal evolution. Provide the mean or median values, the period of time that you are averaging, …
In this paragraph, the authors are just describing the CS values observed throughout the day. Indeed, the hourly averaged CS values are always larger than 0.02 s^{-1}. We compared this value with previous studies over urban and pristine areas.
We modified this sentence into : "Hourly averaged CS values are high (larger than 2 x 10^{-2} s^{-1}) during event days occurring during spring and summer (Figure 8a)."

L334 – Where can we see that?
We did not present any analysis on the undefined events in this manuscript because we were focusing on the conditions favorable for NPF events.

L335, 338 – all the undefined cases show a growth that is stopped? What happen the rest of the percentage?
Yes all undefined events show a growth that stopped. The undefined cases were defined as Dal Maso et al. decision tree suggested. For instance, some particles smaller than 25nm can be observed for more than one hour, but those particles are not growing so the particles remain below 25nm. As the sizes below 15.7nm were not scanned, the particles may have grown from

smaller sizes to 15.7 and then the growth stopped but we do not know about that. These events would be classified as non-event days. For diameters larger than 15.7nm, the undefined days usually exhibit a burst of small particle concentration that disappears during the afternoon as shown in Figure 3b.

A more thorough analysis of these undefined events would be required to investigate the other reasons behind the occurrence of those undefined events. We have not performed that analysis yet and it is beyond the scope of this manuscript.

L341, 342 – almost have not talk about HYSPLIT before, I would introduce here why you use it and the objective. Time, UTC or Local?

As mentioned before, the time is always in UTC. The HYSPLIT local time has been transformed into UTC time. The interest of HYSPLIT was evidenced through the undefined events since we observed that the undefined events are quite often associated with a wind direction shift. Then we introduce HYSPLIT by this sentence: "The shift of the wind orientation leading to a stop of the particle growth indicates that NPF events are associated with certain wind directions or air mass origins. To investigate this, HYSPLIT back trajectories were first sorted as a function of event, non-event and undefined days."

L347, 351 – I am not totally agree with this. CS seems that doesn't play an important role to inhibit the formation of new particles and probably is more the absence of precursor vapors and/or photochemistry (polluted Beijing: Kulmala et al. 2017, Du et al. 2022 or even strong dust events: Nie et al. 2014, Casquero-Vera et al 2020). Are you comparing only clear sky days (no clouds)?

Over ATOLL, the number of dust events was really low but still observable (2-3 dust event per year) and when it occurred, they were clearly characterized by larger $PM_{2.5}$ and PM_{10} concentrations and clearly associated with non-event days.
In these sentences, we wanted to highlight that according to the backtrajectory paths, the air mass could be either enriched or depleted of primary precursors.
The sentence was corrected into: 'Those air masses might then have been slightly enriched in primary precursor vapors. This result is consistent with previous results showing "cleaner" air masses are associated with NPF event cases observed during spring.'

For this analysis, we didn't check the cloud coverage over the whole domain but we observed it at ATOLL. The non-event days, as shown in figure 6a, mostly occur while the cloud coverage is between 0.6 and 0.9. There are a very limited number of non-event (13%) occurring during clear sky (cloud fraction lower than 0.4) conditions throughout the period between 09:00 – 14:00. The study would not have been statistically correct by excluding more than 70% of the data set. We are therefore not comparing only clear sky days.

L373 - NSF6-100 use subindex.
Done.

Section 3.7 – have the authors look the nucleation strength factor for the 50-100 nm size range? It could maybe be an estimation of the increase of CCN due to NPF…??
 As suggested, we plotted the diel profiles of NSF_{50-100} for both seasons (MAM and JJA). The NSF_{50-100} values are ranging from 1 to 1.6. The maximum values are reached during the

afternoon at 16:00 and 15:00 during spring and summer, respectively. Of course, the values are not as important as $NSF_{15.7-100}$ but it still highlights the large impact of the NPF events over ATOLL. Indeed, the concentration of CCN-like particles ($50 < Dp < 100nm$) shows an increase during the early afternoon up to a factor of 0.7 (0.3) during summer (spring). This impact may have a large influence on the CCN concentration available for activation. This point needs to be further studied with a CCN counter to evaluate the hygroscopicity of those particles.

Part of this discussion was added within the manuscript:

"As previously shown(Sebastian et al., 2021), NPF events can also play a major role on Earth's radiative budget when the newly formed particles grow to climate-relevant sizes (50-100nm). In order to understand the NPF influence on these particles the NSF_{50-100} was also calculated (see supplementary figures). The results show a large increase up to 1.6 of the NSF_{50-100} in the early afternoon for both seasons. This suggest a potential impact on the CCN concentration that needs to be further studied."
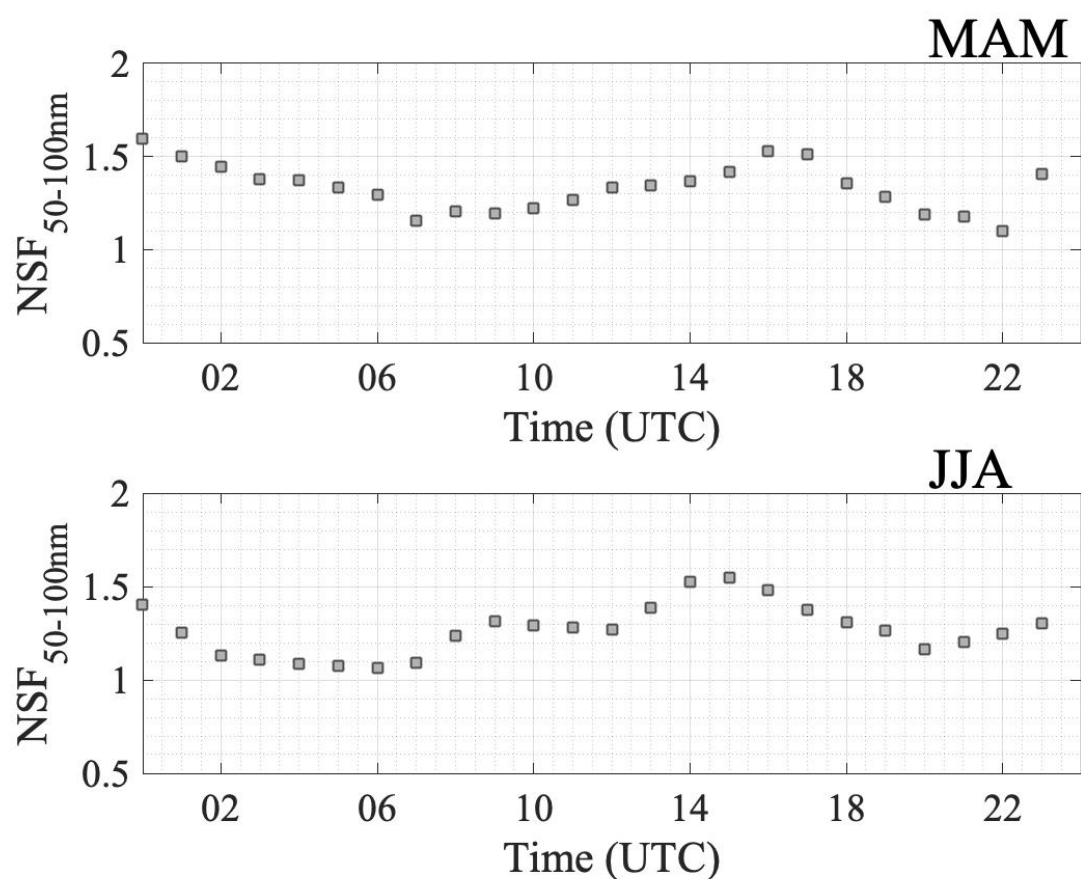

Figure 4: (a) Diel variation of the Nucleation Strength Factor (NSF50-100) during MAM and JJA calculated from number concentration during the 2017-2020 period.

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