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Reply on RC2

Helmi Uusitalo et al.

Author comment on "Occurrence of new particle formation events in Siberian and Finnish boreal forest" by Helmi Uusitalo et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-530-AC2, 2022

We thank the reviewer for the helpful comments and suggestions for improving our manuscript. Below we give our replies (shown in italics) to each of the comments (copied and shown in normal font).

Specific comments:
Objectives: the objectives of the study are not clearly stated. In the introduction the authors state that they seek to understand why "NPF is infrequently observed in Zotino" (line 52), but they fail to properly address it, by limiting the analyses to a display of the observations. However, at line 230 the authors say the "the reasons causing the differences is outside the scope of this paper". Finally, at line 282 it's stated: "The main objective of this work was to find the reasons why…", so the objectives should be clearly stated and appropriate analyses performed and presented to address them. In addition to differences in the instrumentation deployed, both meteorological and chemical conditions should be considered to explore the fundamental sources of discrepancies highlighted (particularly as the issue with the minimum detectable size is not fully provable at ZOTTO and it seems to only partially explain the observed discrepancies).
We will bring out the objectives of the study more clearly and coherently thoughout the text in the revised manuscript. An airmass source area analysis based on backtrajectories and the distribution of biogenic (volatile organic compounds) and anthropogenic (SO2, particulate matter) emission sources in the areas surrounding each measurement site will be added. This will bring information on the meteorological conditions on new-particle formation event and non-event days (the airmass backtrajectories include information on temperature, relative humidity, precipitation and solar radiation along the transport to the site). In the lack of direct measurements of the precursor vapors at all the sites, we can draw some conclusions for the chemical conditions from the comparisons of biogenic and anthropogenic emission sources along the airmass backtrajectories.

Instrumentation differences: at line 53 and through the manuscript the authors suggest that discrepancies in the occurrence of the frequency of NPF events may derive from the too large minimum detectable size of the instrument deployed at ZOTTO. More details about the instrumentation used at the four sites should be provided in addition to the size range covered. For example, details are needed on the number of size bins, temporal resolution, accuracy and uncertainty in the measurements. As observations from different instrumentations are compared, comments about to what extent these measurements are comparable should be included.
We will add more details on the aerosol size-distribution measurements at each site.

Observations time period: details on the four measurement sites should be provided. For example, it is not clear at which heights the analyzed PNSD are measured and if this height is the same at all sites. If there are differences, they should be accounted for (or at least discussed) when comparing the observations at the four sites. Further, it is not clear why observations from 2017-2019 are compared at three sites, while for the ZOTTO site, which is the one showing major discrepancies, data from 2006 to 2009 are analyzed. The different time period analyzed may be a major cause of discrepancies and does not seem to be justified as observations at Hyytiala and Varrio have been collected since 1996 and 1997, respectively. A fair intercomparison of the sites would require at least to analyze the same time period. Since the observations at ZOTTO were collected about 10 years before the data at the other sites, possible changes in the emissions, transport of nucleation precursors may have occurred and should be addressed. All results displayed would be impacted and would need to be revised.
The aerosol measurements at the Finnish SMEAR stations (Hyytiälä and Värriö) and the Russian Tomsk-Fonovaya station are conducted close to the ground (around 4 meters height from the ground). At the ZOTTO site, aerosol measurements are done at two levels: 50 m and 300 m above ground. We will describe in more detail the measurement setup and sampling heights at each site, including discussion on the possible influence on the comparability of the datasets from different sites.
The referee is correct that the different time periods analyzed can cause some of the differences between the sites. The need to use different time periods comes from the data availibility: aerosol measurements were started at Tomsk-Fonovaya in 2011 and in ZOTTO we have only aerosol data available only during 2006-2009. We will add discussion on the possible impacts on our results from these different analyzed time periods, but unfortunately it is not possible to have a common time period including measurement data from all the four sites.

Novelty of the work: The authors should highlight the scientific contribution and novelty of this work. As the methodology is taken from prior studies and the data are not new, more effort should be put into interpreting and analyzing the observations from the four sites in a more consistent and thorough way.
We will sharpen the discussion of our results in the revised version of the manuscript, and we feel that including the modifications highlighted above in response to the referee comments will make the analysis more consistent.

Technical comments:
Line 30: this is an important point as discrepancies in the properties of NPF events could be related to differences in both meteorological conditions and chemical precursors. The literature review should be expanded to include studies beyond boreal forest environments to demonstrate how these variables play a role in dictating NPF occurrence, and how their role varies in different regions of the world. This addition would provide the background for the additional analyses needed to better interpret discrepancies in the data.
Besides those already included in the current version of the manuscript, we will add more references to studies on NPF in other environment types: South African savannah (Hirsikko et al., 2013), rural area (Größ et al., 2018) and urban area (Salma et al., 2021).

Line 99: a classification of NPF events based on a visual inspection of the data is not ideal, as it may be subjective and not reproducible. A quantitative and automatic method to analyze PNSD should be applied.
We recognize the potential subjectiveness of the visual NPF classification method. In order to decrease the influence of this on the classification results, all the NPF classifications are done in groups of several researchers where each member of the group needs to agree whether certain day is classified as NPF event day or non-event day. Otherwise the day is classified as undefined. We think that despite its possible shortcomings, this method of NPF classification still provides trustworthy information on the general patterns of NPF occurrence at each site. Some recent studies have utilized machine learning (ML) methods

in order to automatize the NPF classification (Joutsensaari et al., 2018; Su et al., 2022), however these ML methods still rely on training datasets of NPF classifications which are based on human-made visual inspections of the aerosol size-distribution data.

Line 101: it is not clear how a day is classified as an event day. For how long does a burst of new particles should last and/or for how long does it have to grow and up to which size? These details are needed for reproducibility of the results, particularly as the current event classification is qualitative.
In the NPF event classification, we have followed the criteria by Dal Maso et al. (2005): day is classified as NPF event day, if during the day new mode of nucleation mode particles is observed for more than one hour and the mode diameter grows during the day. We will state these criteria more clearly in our revised manuscript.

Line 113: what is the timestep for each set of observations? If they are different how are the observations homogenized for a proper comparison?
The time resolution of the aerosol size-distribution data is between 10–20 minutes at all the four sites, which in our opinion is similar enough not to affect the formation and growth rate calculations or comparisons of the results from different sites.

Line 131: how is the coagulation coefficient K determined and how is d' p defined?
The coagulation coefficient is calculated according to Fuchs form of the Brownian coagulation coefficient, which is described e.g. by Seinfeld and Pandis (2006). We will add this reference to the revised text.

Line 154: the conversion between nm h -1 and m s -1 is obvious and not needed.
We will remove this detail from the revised text.

Line 181: what are the causes of this nighttime events? Could they be related to some boundary layer dynamics, or specific chemical or transport mechanisms? How do these events look like? Some of the event characteristics may point out to important mechanisms responsible for occurrence of NPF events at ZOTTO and help with the interpretability of the discrepancies with the other sites. These mechanisms should be discussed/interpreted in light of prior literature studies (e.g. Crippa, P., Petäjä, T., Korhonen, H., El Afandi, G. S., and Pryor, S. C.: Evidence of an elevated source of nucleation based on model simulations and data from the NIFTy experiment, Atmos. Chem. Phys., 12, 8021–8036, https://doi.org/10.5194/acp-12-8021-2012, 2012.)
The typical characteristics of ZOTTO nighttime NPF events are described in Section 3.3. of the manuscript. We will add more discussion related to previous observations reported in the literature.

Line 230: basic analyses that would provide more insights and should be added include for example backtrajectory analysis to describe the "climatology" of the events in term of transport of precursors and meteorological conditions, or for specific events to show discrepancies among the sites. Could the NPF events be related to an elevated source or transport of precursors? Sources of discrepancies could be in the instrumentation adopted, but also in the emission context (as the ZOTTO observations are collected 10 years prior the ones at the other sites), availability of precursors (are there any gas phase observations available at some of the sites or from nearby stations?)
As suggested also by Referee #1, we will make additional analyses on the airmass source areas based on backtrajectories. Unfortunately the ZOTTO aerosol size-distribution data is only available clearly earlier than at the other, so the effect of the possible influence from declining anthropogenic emissions (e.g. SO2 and primary emissions of particulate matter) cannot be quantified in the ZOTTO observations.

Figure 6 could be improved as it is confusing to have a different y-axis in the two columns, so the panels are not immediately comparable.

We agree that presenting the size-distributions covering two different size-ranges on equal y-axis scales would make the comparisons between the different panels easier. We will modify Fig. 6 accordingly.

Figure 7: the temporal resolution of the data at ZOTTO is clearly different from the other sites. How is this going to impact NPF detection? As mentioned earlier, for how long a burst of new particles has to grow (and to what size) to be classified as a NPF event? This may also impact the classification at ZOTTO given the different temporal and size resolution.
The temporal resolution of the ZOTTO aerosol size-distribution data is approximately 20 minutes, whereas on the other sites it is 10 minutes. Since the criteria for the appearance time of the newly formed particles is more than one hour, we think that even the sligthly worse time resolution of the ZOTTO aerosol data does not hinder the detection or classification of the NPF events. The greater impact for the NPF event identification in ZOTTO is caused by the higher detection limit (10 nm) compared to the other sites.

Line 260-262: this sentence indicates that no conclusive findings can be inferred from the presented analyses, which therefore need to be expanded to investigate at least key chemical/dynamic processes behind NPF events.
We will expand this section to include results from the additional analyses suggested by the reviewer in this and other comments (airmass source areas and their connection to NPF precursor vapour emission sources).

Figure 9: this figure has a very low quality and should be remade.
We will improve the resolution of Fig. 9

Line 285: here the authors speculate about possible causes, which however should be the main focus of the paper and supported by analyses. A map of the sites may be included when the data are described, along with some meteorological/chemistry/emission summary to help understand the regional background of the events.
We will include in the Conclusions results from the additional analyses suggested by the reviewer in this and other comments (airmass source areas and their connection to NPF precursor vapour emission sources). Figure 1 shows the map of the location of the four measurement sites.

Line 311: the authors seem to suggest the different emission context could be a reason of the discrepancies observed, which would be quite expected. This point should be supported by more data/analyses, as mentioned in previous comments.
We will include here discussion based on the results from the additional analyses suggested by the reviewer in this and other comments (airmass source areas and their connection to NPF precursor vapour emission sources).

Line 318-319: this sentence seems to suggest that the data in ZOTTO are unreliable. More information about the data accuracy should be presented. Have other studies presented/investigated NPF mechanisms at ZOTTO? If so they should be mentioned in the introduction to provide enough background knowledge to the reader. If not, a more thorough analysis of the dataset should be included.
The NPF events at ZOTTO have been analyzed by Wiedensohler et al. (2019) and earlier Heinzenberger et al. (2011) analyzed the general characteristics of the aerosol size-distributions measured at ZOTTO. Both these studies noted the low concentrations of nucleation mode particles. We have mentioned these previous studies in the Introduction.

References:

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