Anonymous Referee #2

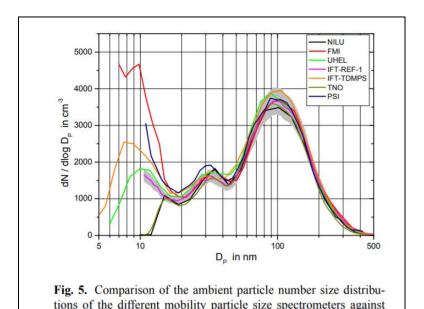
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New particle formation has been demonstrated to play important roles in air quality and climate change. It's essential to classify the new particle formation events and non-events days accurately that can reduce the uncertainty when evaluating the contribution of NPF to aerosol and CCN budget. Previous methods were kind of subjective, and resulted in a poor comparability. This study present an automated method, which is more objective, to classify days into four categories including NPF events, non-events and two classes in between. This automated method was applied in a 10-year NAIS dataset at SMEAR II station. The classification using this methods almost matched the original method, but provided more reliable categories. Therefore, this automated method has the potential to be promoted widely. The manuscript is overall well written C1 ACPD Interactive comment Printer-friendly version Discussion paper and documented. The topic fits well in the scope of ACP. I recommend this manuscript can be published after some revisions.

We thank Referee #2 for their helpful suggestions. We replied to the comments below. The bold text refers to the referee's comments, and the text in italics are additions to the manuscript. The line numbers mentioned in the text below refer to the ACPD version of the manuscript.

1. A NAIS is needed to use this "new" method, which is not easy to be promoted. Can it be used with a SMPS or a DMPS? Hyde have SMPS/DMPS dataset, did the author compare the results that using a NAIS with a SMPS/DMPS? Are they identical?

Our new method uses NAIS in ion mode to account for the process that occurs below 3 nm and represents the early steps of new particle formations which cannot measured by a typical DMPS system (Aalto et al. 2001). While the NAIS is not yet a wide spread instrument, it is important for this study since the ions play a very important role during the early stages of NPF. At these small sizes, the NAIS provides accurate ion concentrations in comparison to DMPS systems which are rather uncertain as seen in the figure below from Wiedensohler et al. (2012). We conclude that, unfortunately, our method is not applicable for data obtained merely from SMPS/DMPS measurements.



shaded area marks the $\pm 10\%$ range around IFT-REF-1. Internal particle losses in all instruments were corrected using the method of equivalent pipe length.

Figure S 1 Comparison of particle number size distributions of different mobility particle spectrometers by Wiedensohler et al. 2012.

the reference mobility particle size spectrometer (IFT-REF-1). The

2. Line 154-156: definition of region events is "initiated over a large area including the measurement location and the particles continue to grow to bigger sizes". Since the SMEAR II station is a surface measurement site, how did the author make sure the identified "regional event" occurred over a large area?

As discussed in a very recent review article (Kerminen et al., 2018), surface measurements at a fixed location can in principle distinguish whether the observed NPF events are regional rather than local in their character. Our classification criteria were designed so that what we call "regional events" would really represent large-scale production of new aerosol particles. The spatial extent of apparently "regional NPF" has been studied in more than 10 individual studies conducted in Europe, North America and China. The general conclusion from these studies is that the spatial extend of regional NPF is typically a few hundreds of km, possibly exceeding 1000 km in some cases (see section 3.1.3 in Kerminen et al., 2018).

3. Transport events, is there any more evidence to support the definition? Any other possibility that other sources but not NPF contribute to the 7-25 nm particle?

Our assumption regarding the transported events comes from the observation of downward flux of particles. One example is shown below by Leino et al. (2018). Interestingly, this is one of many similar events for which we are dedicating a complete separate study including more flight measurements above Hyytiälä (Lampilahti et al. 2019, In Prep).

However, there might for sure be contribution of a different source, for example the time period between midnight and 10 am in the figure below, and which is most possibly attributed to a pollution plume carried to our measurement location and which interferes with our automated method.

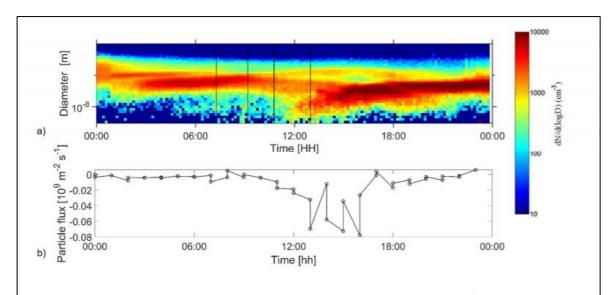


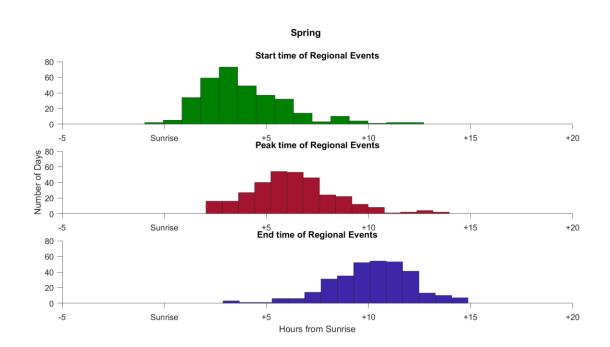
Figure 5. New particle formation event at SMEAR II station in Hyytiälä on 13th August 2015. Panel a) shows the number size distribution measured by Differential Mobility Particle Sizer at ground level inside the forest canopy. Start and end times of two measurement flights were marked by vertical lines in figure. Panel b shows the particle flux measured at 23 m above ground level at the station. Negative particle flux indicates particles flux downwards.

Figure S 2 New particle formation event at SMEAR II station in Hyytiälä on 13th 550 August 2015. From Leino et al. 2018, ACPD.

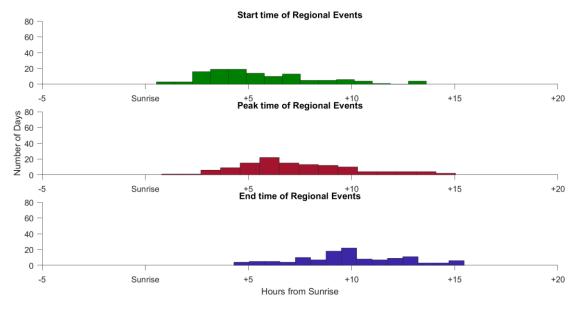
4. Nighttime events: there are some regional events those were started and stopped before sunrise (Fig. 6)? Is it mean they are typical nighttime NPF events? Did they have the "banana" shape? If not, it means these events were not class A event, but still be defined as regional events (see comment 2)?

We thank the reviewer for mentioning this point, we agree that there are a couple of days that have an untypical behavior in comparison to the majority of the days studied. Indeed, we do not observe typical 'bananas' that start during nighttime in Hyytiälä (Buenrostro Mazon et al., 2016; Rose et al., 2018). Our automated method matches the manual classification up to 94% and fails mostly during winter, as shown in Figure S3. The ion concentrations might be disrupted during winter due to snow accumulation. Based on the reviewer's comments we re-considered distributing the start, peak and end times analysis over the seasons, and replaced Figure 6 in the text by the similar one from only spring which is representative of the whole data set and still follows nicely from Figure 5. The results show similarity between all days and spring since the majority of RE occur during spring. Also, the redistribution of the plot into seasons confirms that these events that start and end before sunrise, are indeed in winter. The text in section 3.4 is modified as follows.

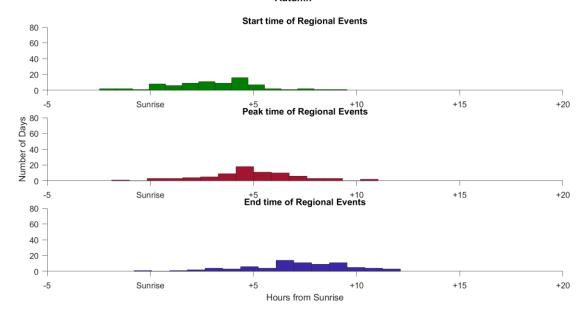
During spring, when most of the NPF events occur, our results (Figure 6) show that indeed RE occur after sunrise and prior to noon, with the maximum number of days occurring between the sunrise and 5 hours past sunrise. The peak times of the events had the most frequent occurrence at 5 to 6 hours after the sunrise, which is between 10:30 and 11:30 local time, complementing our previous assumption that NPF peaks before noon. Finally, the ending times of the events had the most frequent occurrence at 9 to 11 hours after sunrise. During summer the events tend to start, peak and end later than in spring, and they show lower variability in comparison to spring. This observation could be attributed to longer daylight hours and less clouds. Whereas in autumn, the events, start, peak and end earlier than in spring. Exceptionally, during winter, ion concentrations might be affected by the accumulation of snow on or around the inlets. Overall, the variability of the event start, peak and end times can be affected by the solar cycle, degree of cloudiness and seasonality.







Autumn



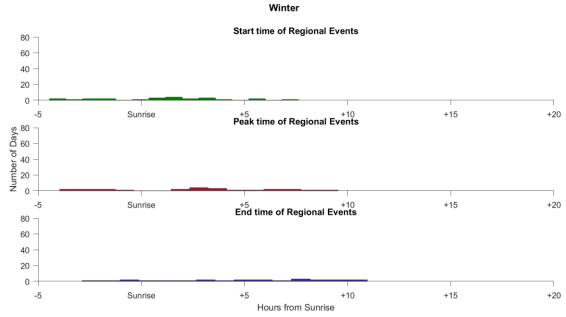
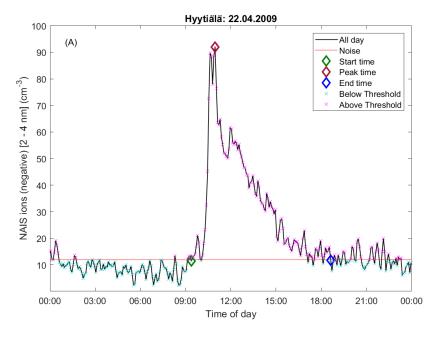
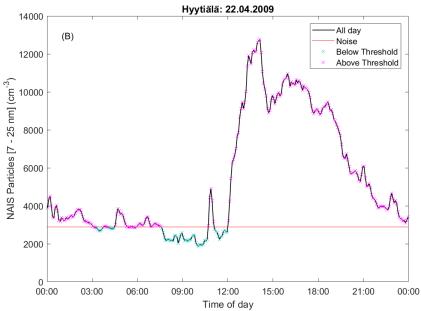


Figure S3. Frequency of days at which regional events start, peak and end past sunrise. For example, most events in Spring start within 5 hours from sunrise.

5. Figure 2: it's better to give an example to show the variation of 2-4 nm particles and 7-25 nm particles in one event.

We agree with the reviewer that showing an example of the variation of ions and particles on the same day improves the quality of our method, accordingly we changed the figures to the same day as shown below.





References

- Buenrostro Mazon, S., Kontkanen, J., Manninen, H. E., Nieminen, T., Kerminen, V.-M., and Kulmala, M.: A long-term comparison of nighttime cluster events and daytime ion formation in a boreal forest, Boreal Env. Res., 21, 242-261, 2016.
- Carnerero, C., Pérez, N., Reche, C., Ealo, M., Titos, G., Lee, H.-K., Eun, H.-R., Park, Y.-H., Dada, L., and Paasonen, P. J. A. C. P. D., https://doi.org/10./acp--173, in review: Vertical and horizontal distribution of regional new particle formation events in Madrid, 2018.
- Hussein, T., Junninen, H., Tunved, P., Kristensson, A., Dal Maso, M., Riipinen, I., Aalto, P., Hansson, H.-C., Swietlicki, E., Kulmala, M. J. A. C., and Physics: Time span and spatial scale of regional new particle formation events over Finland and Southern Sweden, 9, 2009.
- Kerminen, V.-M., Chen, X., Vakkari, V., Petäjä, T., Kulmala, M., and Bianchi, F. J. E. R. L.: Atmospheric new particle formation and growth: review of field observations, 13, 103003, 2018.
- Leino, K., Lampilahti, J., Poutanen, P., Väänänen, R., Manninen, A., Mazon, S. B., Dada, L., Nikandrova, A., Wimmer, D., Aalto, P. P., Ahonen, L. R., Enroth, J., Kangasluoma, J., Keronen, P., Korhonen, F., Laakso, H., Matilainen, T., Siivola, E., Manninen, H. E., Lehtipalo, K., Kerminen, V.-M., Petäjä, T., and Kulmala, M.: Vertical profiles of sub-3 nm particles over the boreal forest Atmos. Chem. Phys. Discuss. (Submitted), 2018.
- Németh, Z., and Salma, I.: Spatial extension of nucleating air masses in the Carpathian Basin, J Atmospheric Chemistry
- Physics, 14, 8841-8848, 2014.
- Rose, C., Zha, Q., Dada, L., Yan, C., Lehtipalo, K., Junninen, H., Mazon, S. B., Jokinen, T., Sarnela, N., Sipilä, M., Petäjä, T., Kerminen, V.-M., Bianchi, F., and Kulmala, M.: Observations of biogenic ion-induced cluster formation in the atmosphere, Science Advances, 4, 10.1126/sciadv.aar5218, 2018.
- Salma, I., Németh, Z., Kerminen, V.-M., Aalto, P., Nieminen, T., Weidinger, T., Molnár, Á., Imre, K., Kulmala, M. J. A. C., and Physics: Regional effect on urban atmospheric nucleation, 16, 8715-8728, 2016.
- Vana, M., Komsaare, K., Hõrrak, U., Mirme, S., Nieminen, T., Kontkanen, J., Manninen, H. E., Petäjä, T., Noe, S. M., and Kulmala, M. J. B. E. R.: Characteristics of new-particle formation at three SMEAR stations, 2016.
- Wiedensohler, A., Birmili, W., Nowak, A., Sonntag, A., Weinhold, K., Merkel, M., Wehner, B., Tuch, T., Pfeifer, S., and Fiebig, M. J. A. M. T.: Mobility particle size spectrometers: harmonization of technical standards and data structure to facilitate high quality long-term observations of atmospheric particle number size distributions, 5, 657-685, 2012.