

Response to Referee #2

The authors thank Referee #2 for time invested in the manuscript. We are happy to hear that Referee #2 accept our response in most parts. In essence, the only remaining larger issue relates to clarification of the effect of seasonal averaging of aerosol number size distributions. In the revised manuscript we have added in Supplementary material Figure S1, a diurnal seasonally averaged size distribution that in addition to the clustering highlight the often observed, intermittent new particle formation events present in the dataset.

We added under 3.1 after first paragraph: “It should be noted that seasonal averages of daily mean aerosol number size distributions do not preserve the signature of new particle formation events (for details regarding new particle formation c.f. e.g. Kulmala et al. 2004). The lack of a distinct nuclei mode in Figure 2 does however not imply that nucleation is absent in the data set, but rather suggest that the intermittent behavior and short lifetime of the nuclei mode under conditions characteristic for the Aspveten station leads to an masking of these features when performing long term averaging. Thus, in Supplementary material, Figure S1, we show Time-of-Day seasonal mean size distributions. As evident, the signature of new particle formation events is present for all seasons except wintertime. 10nm particles are typically observed around noon but grows rapidly into larger size classes during a couple of hours.”

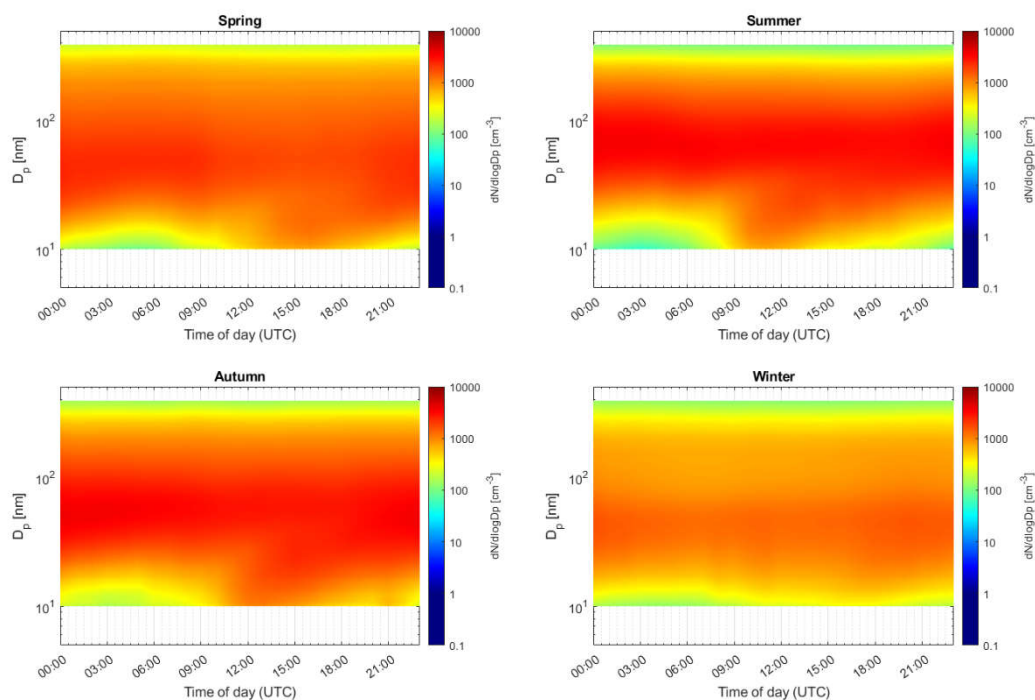


Figure S1: Diurnal variation of seasonally averaged mean number size distributions.

As acknowledged by the Referee, long term averaging over daily means tend to mask the presence of particles in the nuclei mode size range. With aforementioned revision, we hope we have highlighted what might seem as a contradiction depending on how data is presented.

We further expanded the discussion about Figure S3 (Now Fig S4). We have added under Section 4.1, last paragraph: "In figure S4, 95% Confidence interval of Theil-Sens slopes for lower (left) and upper (right) confidence interval. Color indicate calculated linear trend for binned particle number concentration at Aspvreten as particle $\text{cm}^{-3} \text{ year}^{-1}$ for the time period 2000-2017. Areas bounded by the dashed red line represents pairs of month/size bin where test for significance was below the 95% threshold."

Below we provide a point-by-point response to the specific comments raised by the Referee

- 1.) Reference added in revised MS using citation recommended by the reviewer
http://www.climatechange2013.org/images/report/WG1AR5_Chapter08_FINAL.pdf
"Myhre, G., D. Shindell, F.-M. Bréon, W. Collins, J. Fuglestedt, J. Huang, D. Koch, J.-F. Lamarque, D. Lee, B. Mendoza, T. Nakajima, A. Robock, G. Stephens, T. Takemura and H. Zhang, 2013: Anthropogenic and Natural Radiative Forcing. In: Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change [Stocker, T.F., D. Qin, G.-K. Plattner, M. Tignor, S.K. Allen, J. Boschung, A. Nauels, Y. Xia, V. Bex and P.M. Midgley (eds.)]. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA"
- 2.) Changed according to referee suggestion.
- 3.) Now reads: "Seasonal variation of the aerosol number size distribution between 10-390nm presented as daily median aerosol number size distribution for the whole study period, 2000-2017. Superimposed on the surface plot is the median and quartile ranges of integral number concentration."
- 4.) Figure 2 is adjusted according to the suggestion by the referee.
- 5.) Caption now revised and reads: "Figure 2: Seasonally average size distributions observed at Aspvreten 2000-2017. Shaded blue area gives 25th-75th percentile ranges, and dashed line median size distribution. Shown in the sub-frames are same data, but now in log-log scale. Spring=March-May; Summer=June-August; Autumn=September-November; Winter=December-February."
- 6.) Caption now reads "Table 1: Statistics of modal fits per season. Table shows statistics derived from fitted hourly number size distributions. Indicated in table are Nuclei, Aitken and accumulation mode parameters as median and 25th-75th percentile. GSD represents the geometric standard deviation and Dg the geometric mean diameter of each one of the log-normal modes."
- 7.) We added under section 2.3 "The clustering was performed on hourly averaged data, using options "max iterations" of 10000 and "number of replicates" was set to 10 in Matlab." As suggested by the reviewer we also amended first paragraph of 3.2: "This section describes the results from the cluster analysis. As stated under Section 2.3, the clustering was performed on hourly means, roughly 130000 size distributions. This approach captures signature size distributions in different stages of the aerosol life-cycle, including aerosol number size distribution types that originate from new particle formation events (e.g. Kulmala et al. 2004)."

We also point out that in the original MS the second paragraph, section 2.3, reads: "Contrary to standard averaging of number size distributions, cluster analysis and associated centroids can conserve the shape of the aerosol size distribution. Hence, size distribution clusters represent "signature distributions" that reflect contribution from members that are likely to

have undergone similar processing in the atmosphere prior to observations. Thus, clustering size distribution and combining the cluster analysis with auxiliary parameters, such as trajectory derived source areas, temporal distribution of members and parameters related to sink processes (e.g. precipitation) can provide a deeper insight into the multitude of factors defining the aerosol over time. “

- 8.) *Please add a sentence near the start of this section re-iterating this point about the additional value/virtue within the cluster analysis technique, that (even if this is not yet explored in this particular manuscript) the approach potentially enables to identify changes in nucleation within the long-term measurement record that exists at Aspreveten and other sites.*

In Section 2.3 and in revised section 3.2 we address this, but we do however agree that this could be re-iterated. We thus add in Discussion: “In addition, cluster analysis of hourly number size distributions has been demonstrated to be a useful tool in trend studies. The method has been applied to study how the aerosol observations are distributed over 12 signature distributions, and further applied to investigate how the aerosol have been re-distributed between these 12 dominating cluster types during the period 2000-2017. The method seems well suited for studying trends in new particle formation events.”

- 9.) First bullet in conclusion reads: “As revealed by the aerosol size distribution clustering, it is evident that the cluster representing clean, cloud processed aerosol is increasing on expense of the polluted type monomodal size distribution. There is only marginal increasing trend of cluster members belonging to clusters showing sign of recent new particle formation. At the same time, the most polluted cluster 12 has been reduced from around 5% of observations to around 1% during the period 2000-2017.”

We add to this bullet

“We have shown that cluster analysis successfully can be used to study aerosol trends, and further that cluster analysis also can be used for studying trends of intermittent processes such as new particle formation events. The method has clear advantages compared to standard time averaging techniques as it preserves the shape and number concentration of the aerosol number size distributions within the clusters which otherwise easily can get lost in usually applied time averaging such as mean and medians. “