

The paper by Sulo and co-workers presents long term measurements of sub-3 nm particle concentration and their precursors conducted in Hyytiälä, in the boreal forest. The first part of the study focuses on the identification of optimal settings of the PSM (used for particle measurement) for this site. The second part is dedicated to the study of the time series, including diurnal cycles, of the gas and particle concentrations. The involvement of the selected vapours in the formation of sub-3 nm particles is finally addressed in a last part by the mean of a correlation analysis.

While the data set used is of undeniable value and the objectives presented are of obvious interest, I am however reserved on certain aspects of this study. My main concern is about the definition of the particle size classes used for the analysis, which seem to me too fine in view of the uncertainties associated with the measurement, with a probable impact on the results presented, and in particular on the correlations. Moreover, it is sometimes difficult to extract the main messages from the second part of the study, which is very descriptive, and which I believe would benefit from being sometimes more synthetic. The integration of a more "chemical" dimension to the analysis proposed in Sect. 3.2 would finally, in my opinion, make this second part more complete. These different aspects are presented in more detail in the comments listed below, which I think should be considered before publication of this work.

P2, Introduction: measurements performed in Hyytiälä have enabled numerous studies to be carried out, in particular on the understanding of new particle formation and the identification of its precursors. I would thus suggest to include in the introduction a paragraph recalling some key results specific to this site in order to better situate the objectives and interest of this new study in relation to past work.

P6, L177: The authors indicate the appropriate settings for the station of interest but it is not completely clear to me to which extent these settings are site specific. Could the authors add a sentence or two to briefly comment on these aspects, and discuss in particular the possibility of extrapolating the results obtained to other sites, under what conditions?

P7, Measurement uncertainties: measurement uncertainties related to the nature of the particles and sampling conditions have been the subject of various studies in recent years and are clearly recalled here. Given these uncertainties, I wonder what is the relevance of size classes as fine as those proposed in this work. In particular, the width of the proposed bins is of the same order or less than the uncertainty related to the chemical composition of the particles or their charge. My interrogations are reinforced by the fact that on NPF event days (Fig. 5), the evolution of the concentrations does not seem to show any growth link between the different classes, or at least between the 2 last ones which are considered to be more connected to NPF. I think it would therefore be more appropriate to reduce the number of classes.

P10, L254-256: is the frequency of events of marked stratification known, significant? Should the correlation analysis reported in Sect. 3.3 be limited to day time in the "All data" cases?

P11, L280-282: "Correlations were also separately investigated for spring- and summertime NPF events. There were not enough data points for events during autumn and winter for separate analysis during those seasons." If I am not mistaken, there are correlations reported for autumn in Table 3.

P11, L286: Could the authors add a few words on the value of distinguishing between nitrates and non-nitrates?

P12-13: Time series of the particle concentration:

- L320-321: "We observe a clear annual maximum during late spring and early summer". I would say that this statement is too strong since it seems to me that it is only verified for 2 years (2016 and 2018). In 2015 and 2019, the concentrations measured in autumn are of the same order as those measured during late spring / early summer, and in 2017, despite the lack of data, it seems that the autumn levels are even higher than those of the previous months.

- L324-332: “Excluding this part of the data did not have a significant impact on the rest of the analysis.” Does this mean that the data were effectively excluded for the rest of the analysis?
- L336-337: “and because their data was not filtered to remove scans with too high background”. The difference in concentration between the two studies is relatively large (almost an order of magnitude), and I am not sure that the proposed hypotheses can explain such differences. In particular, Fig. 1 suggests that scans with a high background are not systematically associated with higher concentrations than those associated with lower backgrounds (or is it only true for the smallest particles, i.e. in the class 1.1-1.3 nm ?). Was the background itself subtracted from the data in Kontkanen et al. (2017)? Also, I think that based on the studies by Lehtipalo et al. (2014) and Cai et al. (2019), it cannot be excluded that the use of methods other than Kernel (e.g. step wise) could have contributed to the observed differences as well; however, unless I am mistaken, the inversion method used by Kontkanen and co-workers is not specified in their paper.

P14-15: diurnal cycles of the cluster concentration

- The use of a logarithmic scale makes the identification of certain maxima / minima very difficult!
- L369-372: I do not think that the peak observed on event days around noon in the size range 1.7-2.5 nm can be described as a strong maximum. On the other hand, the link between these observations and the occurrence of regional NPF events does not seem obvious to me either; I would expect in this case a chronology in the increase of concentrations (i.e 1.3-1.7 nm and then 1.7-2.5 nm) that is not seen here. Is it also possible that this unexpected chronology is linked to measurement uncertainties / definition of the size classes?

P16: diurnal cycles of the cluster concentration in each season

- The end of this section is very descriptive, with observations that are often difficult to confirm due to the logarithmic scale (e.g. L389), and it is globally difficult for me to extract a message from this analysis. I would suggest to add one or two sentences at the end of the section to summarize the main outcomes.
- Concentration levels and the presence or absence of distinctive diurnal cycles are primarily related to the frequency of occurrence of NPF in each season. However, this explanation does not seem to be sufficient, since although the frequencies of NPF are lowest in winter, the concentrations observed are on average higher than those in autumn for the 2 size classes 1.3-1.7 and 1.7-2.5 nm, and comparable or even higher than those in summer for the class 1.3-1.7 nm.

P17-20: Time series of vapour concentrations:

- This section is once again very descriptive, with an analysis that sometimes seems too superficial to me. It would, in my opinion, benefit from being more detailed from a chemical point of view, particularly with regard to the processes involved in the formation/transformation of the organic compounds of interest according to their nature (monomers vs. dimers, nitrates vs. non-nitrates) and their influence on the cycles observed. The findings of studies previously dedicated to the specific analysis of cluster composition at the site (e.g. Yan et al. 2016; Bianchi et al., 2017; Rose et al. 2018 and connected literature) could for instance be explicitly mentioned to benefit the interpretation of the results.

- L406: “during summer months”: based on Fig. 7, it should in my opinion be changed to “between late spring and early autumn”.
- L445-450: The importance of SA and HOM non-nitrate monomers in the formation and initial growth of the clusters cannot be deduced with such level of confidence from the comparison of median diurnal cycles alone (“This points to the importance”, “implying that”). Based on Fig. 5, the other selected compounds also have variations that are comparable to that of the cluster concentration, and could thus be involved in their formation as well (HOM nitrate monomers peaking around midday, dimers peaking in the evening). I would suggest using more moderate phrasing at this stage of the analysis.
- L451-487: As in the previous section, I find it difficult to extract the main messages from this relatively long description. I would suggest, for example, to first highlight the fact that the general pattern of the cycles is overall comparable for all seasons, but with amplitudes and/or periods that are variable and probably modulated by the amount of global radiation; I would then describe the main differences observed between each season.

P21-25: connection between precursor vapours and cluster concentrations

The search for correlations between the presence of newly formed particles and possible precursors seems to me relevant for the identification of the compounds involved in the formation and early growth process. However, I believe that the uncertainties related to the definition of such fine particle size classes as those used here limit the relevance of such correlation study, whose results are for me difficult to interpret. I still have a few comments on this section:

- L507: I would suggest to better explain the choice of the combinations of compounds to be investigated.
- L515: “However, the difference in correlation coefficients is not large”: It depends on the meaning of large but I would say that the difference is sometimes significant!
- L520-521: “However, the correlation can also point to two separate formation pathways, organic and inorganic.” I would suggest to mention the earlier results of Yan et al. (2018) to support the idea of multiple mechanisms.
- Fig. 10: Line 494-495 indicates that calibration coefficients are not considered in the correlation analysis, but they seem to be accounted for in Fig. 10. Furthermore, the orders of magnitude shown on the x-axis in the third column (product of the previous 2 columns) do not seem to be consistent with what is shown in the previous columns.

P25-26 : Conclusions

Beyond the fact that it remains difficult to approve certain observations due to the use of logarithmic scales, some of the information that is recalled here does not seem to me to be precise enough, or not in line with what is reported in the previous sections:

- L592: “The 1.3–1.7 nm and 1.7–2.5 nm particle concentrations show a marked increase during springtime”: this is not obvious for 1.7-2.5 nm particles, whose concentration is close to what is observed in summer.

- L593-594: “The diurnal patterns of sub-3nm concentrations exhibit clear daytime maxima around midday.”: It does not seem to be true for all sizes, seasons, types of days (NPF vs non event days).
- L594: “This maximum is the clearest during spring and autumn”: Barring any misunderstanding on my part, this message seems to me to contradict what is reported in L393-396 regarding autumn: “weak diurnal pattern”, “no discernible diurnal pattern”.

Technical / Minor comments

P2, L43: point missing at the end of the sentence.

P3, L79: “for measuring particle concentrations larger than 1 nm in size”: I would suggest to change to “for measuring the concentration of particles larger than 1 nm in size”.

P5, L144: extra point at the end of the sentence.

P11, L278: extra “then”.

P13, L334: “concentration” should be removed at the end of the sentence.

P19, L460: point missing at the end of the sentence.

P24, L558: “correlations” should be removed.

Figures:

- I would suggest homogenising, as far as possible, the appearance of the figures and in particular the font size used. Adding a grid would also make the values easier to read.
- Fig. 5: the scale of the ordinate axis (left) of the third panel should be slightly adjusted to match the scale of the others and ease the comparison.

The writing of times should be harmonized (e.g. L389 vs L424).