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Interactive comment on "Factors influencing the contribution of ion-induced nucleation in a boreal forest, Finland" *by* S. Gagné et al.

Anonymous Referee #3

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This article contains interesting experimental material on the role of ion-induced nucleation, as opposed to the nucleation of neutral molecular clusters. Important assets are the extensive data base and the various approaches used with regard to the data evaluation. The data and ideas are very much original, so that the paper appears suitable, after revisions, for publication in ACP. Several revisions, however, appear very necessary. My concerns and suggestions are outlined below.

A first general observation concerns the structure of the results Section 3. Here, the authors plunge into the results by showing a rather abstract Figure 1, which represents quite advanced yet abstract results obtained after much data processing. While the Figure itself is fine, it is a quite unfavorable start for a results section and should appear much later, i.e. when the reader has been faced in a more concrete manner with

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the experimental data. Once concrete examples have established a solid imagination of your data base, you can easily switch to the more complex and statistically-based results.

Therefore, in view of the general importance of the article as well as the rich data base available I would appreciate to see a few selected case studies first, that help the reader to see clearly what is going on in terms of particle numbers (charged, non-charged), gaseous precursor and micro-meteorological parameters. Diurnal patterns of these parameters shown simultaneously in the same graph would help to see what is going on. In the conclusions, we can later read that "both ion-induced and neutral nucleation are taking place in the same event" albeit in different stages of the event evolution. I strongly assume that you may find some illustrative examples to support this conclusion from the beginning of the results section. This would help a reader appreciate that your conclusions are real.

A second comment concerns the Section 2.2.1 (data classification). This section, which puts the shape and relevance of all subsequent results on the map, appears rather superficial, and remains only partly comprehensible for a reader even after studying the references Dal Maso et al. (2005) and Gagné et al. (2008) cited. To enhance traceability, and enable other researchers a comparison with their results, this section should be significantly expanded and clarified. Enhancement of this section is vital also in view of certain amounts of data having been removed from further analysis. See also comments below.

Third, in Section 3 you often write about significant or insignificant differences in terms of aerosol and other atmospheric aerosol parameters between overcharged/undercharged days. However, "significance" is not well defined in your article. Since this aspect of significance or insignificance of differences leads to the main conclusions of the article, it is advisable to introduce a quantitative measure whether the observed differences are of importance of not for the charging state of the nucleation particles. For instance, you could help yourself with statistical tests of various kinds. It would be great if a Table could summarize the results of such statistical tests, accompanying the text blocks in Section 3.2.

Fourth, your interest is focussed on the charging state of nucleation mode particles. I would also be curious to learn about differences in the concentrations of charged/non-charged particles in the bigger size ranges? Do they correspond to what we expect, i.e. how close are they to Boltzmann's charge equilibrium?

In the conclusions, I am also missing a statement on how relevant ion-induced nucleation might be, after all, in the boundary layer. How much is it likely to contribute to the average number of 3, 10 nm particles?

Detailed comments:

"overcharged days", "undercharged days": these terms sounds very much like technical jargon at the very beginning of the abstract. Define these terms first, and reformulate in a more general language.

p. 25804, I.11: bipolar charger can be "switched on and off". A radioactive charger cannot be switched on and off, it's always "on". Probably you want to say something like "the bipolar charger can be bypassed", or "aerosol samples can be conducted either passing through or around the bipolar charger".

p. 25804, I.23: "when the charging state is smaller than the one": sentence hard to understand.

Section 2.2.1, data classification This section is the part of the paper that essentially defines the data set and subsequent analyses. However, it appears quite superficial in its present form. The methods of event selection and classification are drafted only very briefly, although they are necessary, for instance, for other researchers to compare the results quantitatively with their own work. In summary, I recommend the entire section to be expanded and clarified. If necessary, Figures could be added to illustrate and oppose clear and ambiguous cases of charged, undercharged, and steady-state

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events.

"loosely based on the classification of Dal Maso et al. (2005)": Even if the classification is only "loosely based" on the previous work, some sentences should be added to clarify the procedure itself to the more ingenuous reader, and illustrate the differences to Dal Maso's work, particularly when event frequencies are compared.

p. 25811:, l. 16: "thermodynamic principles": What are these principles and how do they relate to the present results?

p. 25811:, I. 20 and Figure 2b: "the agreement is good". How do you arrive at this conclusion? Sincerely, I wouldn't call differences by a factor up to five in both directions a "good agreement". It might be advisable to add bars of uncertainty to the data points and scrutinize the reason for the many "outliers". In fact, this comparison suggests that one of the methods to determine growth rates is questionable.

"External radiation": If the external radiation considered is gamma radiation, why not calling Section 3.2.4 "Gamma radiation"?

Section 3.2.5 "Growth rates" The results are only briefly stated. What I am missing here is some discussion about what your findings are relevant for, and whether they are in line with theoretical expectations.

Fig. 2b, caption "Ion induced fraction" of what?

Sloppy language, should be improved stylistically.

Sect. 3 "when the events are going on" "a bit below 10 ion pairs" (exactly?) "simple" (simplistic)

Sect. 5 "steal away" "perhaps"

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