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Atmospheric Chemistry and Physics Discussions

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

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Answers to the interactive comment on "Factors influencing the contribution of ion-induced nucleation in a boreal forest, Finland" by S. Gagné et al. Anonymous Referee 2 Received and published: 5 March 2010

First of all, the authors thank the referees for their comments that have improved the clarity and the quality of the manuscript. Answers below.

Overall comment: The paper describes a vast set of field site measurement data on ultrafine particles. The particles have been measured by a DMPS instrument with and without a radioactive neutralizer, obtaining fairly high time resolution data on the electrical charging state of the particles. The IIN vs. neutral nucleation is concluded to

vary with the temperature, solar radiation and humidity. The dataset and the conclusions which have been made here should be published, but this manuscript should be slightly improved to meet the quality of the dataset itself.

Specific comments:

Why were the days when different polarities did not agree, discarded? I would assume that appearance of the case one polarity being overcharged and the other undercharged, would give exceptional information of the unbalance of the IIN of different polarity. The whole assumption is that the particles remain charged after being nucleated by IIN. And, did the discarded days have temperature difference in respect with any other group of data?

We discarded those days to make clear categories of higher IIN and lower to inexistant IIN. Of the 29 days, 28 were slightly overcharged for one polarity and at steady state for the other. These events would be in a "middle" category between overcharged and undercharged, and would not help to distinguish the features that have an effect on Ion-induced nucleation of neutral nucleation. We felt that it was better to have two clear categories rather than making on of those two categories "weaker" or having a third category.

We looked at the temperatures for these 29 days and the temperature of this group is in between the temperatures of both categories. (see Figure 1). Here the red line is the overcharged median, the blue line the undercharged median and the full black line is the discarded based on polarity disagreement median. The shadowed area and the black dashed lines show the 25th and 75th percentiles with the corresponding colors. As expected, those discarded days are closer to undercharged days because they were all (but one) weak overcharged + steady-state. When we include the summer months, the temperature lies right in between the overcharged and undercharged temperature curves (not shown here).

We agree however that a day when one polarity is overcharged while the other is undercharged would be particularly interesting, especially if a mass spectrometer would participate in the measurement campaign. Unfortunately, in the period analyzed in this study, only one such day was found (in March 2006) and no aerosol mass spectrometer was in Hyytiälä during that period. We added more explanation about the reasons behind this choice in the manuscript at the end of section 2.2.1.

“Of these 29 days, only one case of overcharged negative polarity and undercharged positive polarity was observed, indicating that both polarities had different chemical pathways in accordance with Eisele et al., 2006. All the other days showed a weak overcharging for one polarity and steady-state for the other one. These days are not uninteresting but they are hard to classify with respect to “little or no IIN” which is called undercharged in this study and “higher contribution of IIN” which is called overcharged in this study. They are cases in between, thus, in order to make the differences between the parameters easier the observe, those gray cases were discarded.”

Row 358 [p.25815 line 12] states that ‘NAIS data came only from spring days’. The reader is confused. So, NAIS was not measuring the whole period of 2 years 7 months? Please, make a clear summary of the measurement periods for different instruments.

The NAIS was in Hyytiälä only during this period: 6.3. - 16.5.2006, 14.9. - 15.12.2006, and 8.3. - 27.8.2007. (Manninen et al., 2009, BER). This period was added in the instrument description section 2.1.4

“During the period covered in this study, the NAIS was in operation between 9 March and 16 May 2006, between 15 September and 12 December 2006 and between 8 March and 27 August 2007.”

We also clarified the reasons why the formation rates are only available for spring in the manuscript in section 3.2.4 just before the sentence mentionned by the Referee:

“Although the NAIS was also measuring during other seasons, the formation rates were available only for days during spring. This is because formation rates are most reliable

only for the strongest NPF events, which happened to all take place in spring, in this study.”

The temperature averages of the event days differ for overcharged and undercharged, as shown by Figs 3a and 3b. But, also the time of year i.e. the seasons when they occur, are different. Please specify, again, which fraction is actually assumed to be driven by the temperature and which fraction by the season. Can the method of Fig 3b be repeated for solar radiation and relative humidity as well? Frankly speaking, I’m not fully convinced of the plain temperature difference being the main reason, just based on Fig 3b. Isn’t the seasonal variation still somewhat ‘built in’ in the data presented in Fig 3b, although the (arithmetic) delta T is considered? Undercharged events do occur on seasons which tend to be colder, right? To my opinion, it should be stated already in the abstract that yes, IIN was higher on warmer and sunnier days, but also a clear seasonal difference was found, even slightly updated from results by Gagne et al Tellus 2008.

We have calculated the temperature difference for all days (not shown but the difference, 8.5°C, is stated in figure 3a’s caption), then for all non-summer (fig. 3a) days, and then the seasonal temperature difference (fig 3b). We do not feel like the seasonal variation is still “built in” the data. For each day the delta T is the difference between e.g. T(1 March 2006) and mean(T(1 March 1996:2008)), so that the day is colder or warmer than the average days at the same time of the year.

Our hypothesis is supported by models e.g. that of Lovejoy et al., 2004 (in this case a sulfuric acid and water model). “Low temperatures, high RH, and high [H2SO4] decrease the height of the barrier and facilitate nucleation.” And this barrier is very small or even inexistant in the case of ion-induced nucleation while it exists for neutral nucleation (see Lovejoy et al., 2004, figure 1). However, we agree that it is difficult to quantify the part that is due to the conditions that exist in summer and the part is due to the temperature alone based on the dataset available. Chamber experiments with controlled parameters would probably yield clearer results. We added comments on the seasonal trend in the conclusions to make sure that both influences will be considered by readers.

Lovejoy, E.R., Curtius, J. and Froyd, K.D.: Atmospheric ion-induced nucleation of sulfuric acid and water, J. Geophys. Res., 109, D08204, doi:10.1029/2003JD004460, 2004.

These sentences were added to the conclusions "We showed that, in Hyytiälä, days with a bigger fraction of ion-induced nucleation tended to occur on warmer, dryer (lower relative humidity), and sunnier days and more often during the summer time." and "Due to the clear seasonal trend, there are probably other factors that promote ion-induced nucleation or limit neutral nucleation, for example an increase in abundance of volatile compounds in summer."

The authors would like to point out this section of the manuscript too (p. 25818, lines 19-22): "The magnitude of this effect is unfortunately hard to assess quantitatively as the molecular-level mechanisms for both neutral and ion-induced nucleation are, as yet, unknown. Other parameters, such as seasonally dependent volatile organic compound emission, might also influence the nucleation mechanism."

We applied the same method to relative humidity (RH) and solar radiation, and we found that the relative humidity had a median difference of -17.52

The same method was also applied to global radiation. The median difference for overcharged days was 64.73 Wm-2 and 35.53 for undercharged days. This means that event days happen on sunnier days than non-event days, and also that overcharged days happen on sunnier days than undercharged days. See figure 3 (where GR stands for global radiation).

We added these numbers in the text in section 3.2.1.

"Using the method shown in Fig. 3b, we got a median difference in relative humidity of -17.5

"Using the same method as in Fig. 3b, we got a median difference of 65.7 and 35.5 Wm-2 for overcharged and undercharged days respectively compared to the mean of the previous years at the same time of the year."

Minor comments: Row 108 supposed to read: "...two plain aspiration-type DMAs"

Yes, this was corrected.

Rows 356- 358 [p.25815 line 12] Unclear sentence ending with 'that'.

The authors are not certain what version of the manuscript this is. We could not find this in the ACPD typsetted version of the manuscript. We also asked the typesetter to remove the "that" at the end of the sentence before it was available on ACPD. We will make sure that it is corrected in the next version.

Row 587 [p.25824 line 15] McMurry, P.H. Row 573 [page 25825, line 1] McMurry, P.H.

Yes, this was corrected.

Row 692 [Table 1 caption] supposed to read "... new particle formation events. "

Yes, this was corrected.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 25799, 2009.

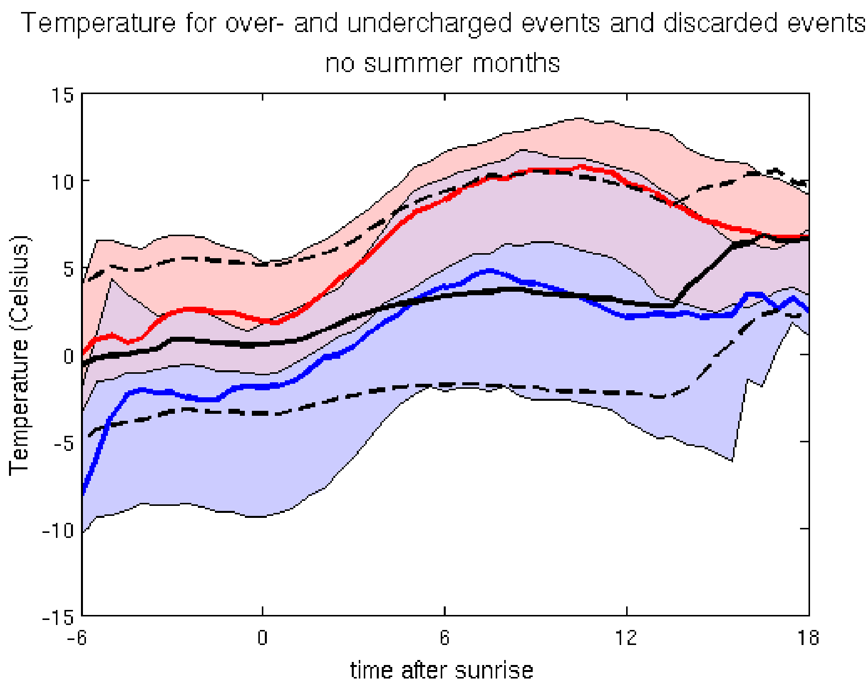

Fig. 1. Figure 1 - Temperature for the discarded days (in black) compared to over- and under-charged classes.

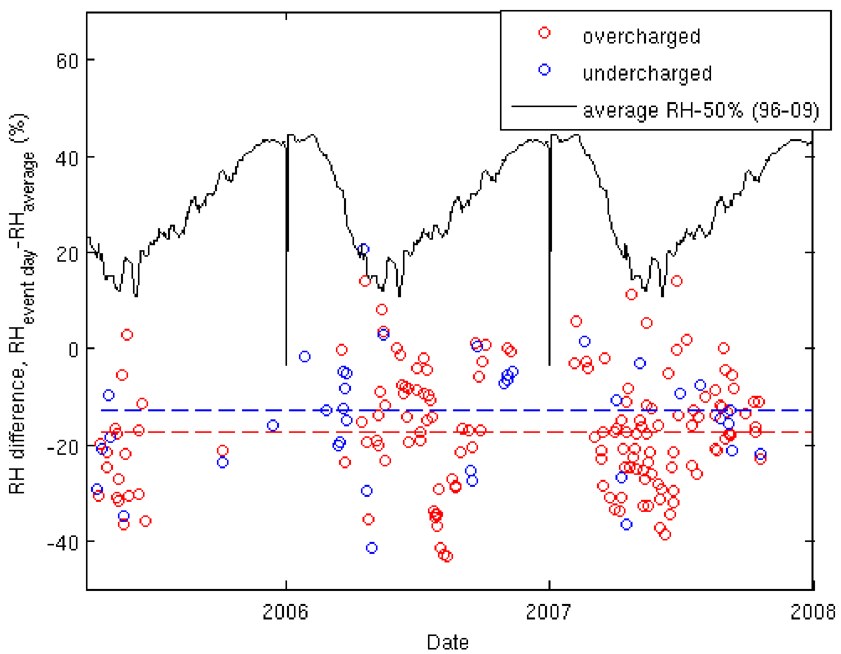

Fig. 2. Relative humidity difference between the event day and the average of the past years on the same day.

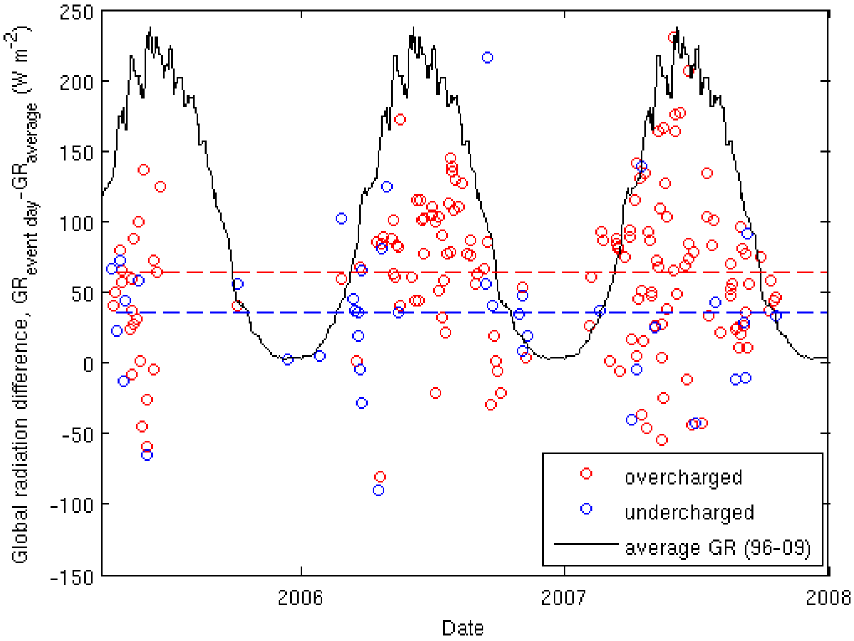

Fig. 3. Global radiation difference between the event day and the average of the past years on the same day.