Atmos. Chem. Phys. Discuss., 9, C11534–C11538, 2010
www.atmos-chem-phys-discuss.net/9/C11534/2010/

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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Received and published: 15 March 2010

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 #1 Received and published: 6 December 2009

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

General Comment Authors deserve praise for their persistent efforts over many years on understanding the contribution of the IIN to the NPF in the atmosphere. There is no doubt that authors are knowledgeable on this subject, and they provide meaningful discussions on their experimental results. Authors provide convincing interpretations

on the observed difference in the temperature and H2SO4 between overcharged and undercharged events. Hopefully in the future, authors use their computational chemistry model and quantify the probability of IIN and neutral nucleation and corresponding energy barrier under the measurement condition.

I list my comments below.

Page 25800, line 14-18. These last two statements are vague. The word "tentative" seems unnecessary. It is better to mention "neutral nucleation and IIN" rather than "different nucleation mechanisms". It would be more effective to state some specific ideas the author would like to propose the most.

Yes, we modified the abstract according to these comments and added this sentence at the end of the abstract.

"For example, we propose that these observed differences could be due to high temperature and low relative humidity increasing the height of the energy barrier a particle has to reach before it can grow and thus limit neutral nucleation."

Page 25803, line 11. It is helpful to add the half life of Ni-63, so that the activity level does not change significantly over 23 year. It is critical to re-state the measurement size range of the Ion-DMPS.

Yes, the half-life is about 100years and the measurement range was 3-15 and 2-11.5 nm depending on the period. This information was added in sections 2.1.1.

"The Ion-DMPS (Mäkelä et al., 2003; Laakso et al., 2007) is based on a Differential Mobility Particle Sizer (DMPS, Hoppel, 1987; Aalto et al., 2004) whose bi-polar charger (Ni-63, 370 Mbq, half-life of ca. 100 years) can be switched on and off and whose Differential Mobility Analyzer (DMA, Winklmayer et al., 1991) can classify particles of positive and negative polarity according to their electrical mobility. The size range covered was from 3.0 nm to 15 nm mobility equivalent diameter between April 2005 until mid December 2006 and from 2.0 nm to 11.5 nm after that."

ACPD

9, C11534–C11538, 2010

Page 25807, line 713. It is unclear why both polarities need to be overcharged for them to be categorized as "overcharged". The chemical composition of initial nuclei are different between positive and negative IIN (Eisele et al. 2006). IIN can occur on one polarity or both polarities. It seems inconsistent that authors considered the NPF event having the charged fraction being at steady state as "undercharged" although authors are aware of the memory effects on the charged fraction during the particle growth. We do not know whether the neutral nucleation or ion-induced nucleation was the dominant nucleation mechanism.

Eisele, F., Lovejoy, E. R., Kosciuch, E., Moore, K. F., Mauldin III, R. L., Smith, J. N., McMurry, P. H. and Iida, K. (2006). Negative atmospheric ions and their potential role in ion-induced nucleation. J. Geophys. Res. 111:D04305/04301-D04305/04311.

Yes, it would be interesting to do the analysis separately for both polarities, especially in combination with a mass spectrometer (which became available in Hyytiälä only later). However, in this study, the vast majority of the days showed the same charging behavior for both polarities, although this method does not provide information about the chemical compounds participating in negative and positive particle formation, it does however provide information on the influence of the electrical charge in relation to neutral pathways.

As we explained on p. 25807, lines 8-11, 40 days were discarded. 29 were in a case where one polarity was differently classified from the other. Of those 29, 28 had a combination of a weak overcharging and a steady-state. Only one day (March 2006) was classified over/under. That day would indeed be extremely interesting to study with regard to the underlying chemistry. Unfortunately, there was no aerosol mass spectrometer on the site on this day. Such information would have been particularly interesting. We added more explanation about the reasons behind this choice in the manuscript in section 2.2.1 and the reference was added. "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

ACPD

9, C11534–C11538, 2010

with Eisele et al., 2006."

The classification of "steady-state" events as undercharged was only to show that the charging state was fairly small as explained at the end of section 2.2.1. By using such a separation method, days with a significant (but not dominating) amount of IIN are separated from days with less IIN. Moreover, with the values of growth rates observed in Hyytiälä, the memory should be preserved in most cases. Also, given the higher growth rates on undercharged (and steady-state) days, as explained in section 3.2.5, it is unlikely that steady-state events are overcharged events that lost the charge information.We modified slightly the explanation of section 2.2.1 in an attempt to make it more intelligible.

Page 25811, Figure 2b. Readers would like if the values on the vertical and horizontal axis are switched. It is more conventional that the true values, which are NAIS, are given along the horizontal axis.

Although the authors don't think the NAIS gives absolutely "true" values, we agree that it may be less sensitive than the fit from Kerminen et al., 2007 and more commonly used, hence we have changed the axis as recommended. The NAIS formation rates ratio method including uncertainties, especially in the 2-3nm size range. The method is described in detail in: Manninen, H. E., Petäjä, T., Asmi, E., Riipinen, I., Nieminen, T., Mikkilä, J., Hõrrak, U., Mirme, A., Mirme, S., Laakso, L., Kerminen, V.-M. Kulmala, M.: Long-term field measurements of charged and neutral clusters using Neutral cluster and Air Ion Spectrometer (NAIS). Boreal Env. Res. 14, 591–605, 2009b.

Page 25818, line 23-. It is recommended that authors investigate whether the formation rate under positive or negative IIN is proportional to either closer to $H2SO4^1$ or $H2SO4^2$. Has anyone show this analysis before using field data? Authors seem to be the only research group that has large enough data set to do such analysis.

This question is currently being investigated and is going to be addressed in a coming publication (Paasonen et al., manuscript in preparation).

ACPD
9, C11534–C11538, 2010

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

ACPD

9, C11534–C11538, 2010

Interactive Comment