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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Identification and classification of the formation of intermediate ions measured in boreal forest" *by* A. Hirsikko et al.

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In this manuscript, the authors presented a statistical summary of event, undefined, and non-event days with regard to intermediate ion (or aerosol) formation, based on three years of the Balance Scanning Mobility Analyzer (BSMA) measurements of the size distributions of air ions (0.42 - 7.5 nm). The authors concluded that the number of days in different classes based on BSMA measurements differed slightly with their earlier categorizations derived from Differential Mobility Particle Sizer (DMPS) measurements. The authors further divided the nucleation event days (class I) into three sub-classes (Ia, Ib1, and Ib2) based on the properties of the observed mobility distributions. Analysis of the difference between charge polarities indicates that the negatively charged particle formation were generally more favorable under the conditions at the

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measurement site. The authors also reported the measurements of rain-induced intermediate ion bursts and the appearance of intermediate ions during snowfall. This is an interesting paper and the results and analyses presented in this paper are useful to improve our understanding of the particle formation mechanism. I think that the paper can be further improved to make it clearer and probably more significant. My suggestions/comments are given below.

Specific comments:

1. The evolutions of charged clusters range from molecular sizes to around 7.5 nm as measured by BSMA provide useful information to identify the possible nucleation mechanisms, especially about the role of ions in nucleation. With regard to nucleation mechanisms, the authors pointed out: (1) The intermediate ions are either formed via ion-induced nucleation or via the attachment of small ions on neutrally nucleated particles (page 9193, lines 1-3). (2) The gap in the size distributions of cluster ions (subclass Ib.2) is an indication of the dominance of the neutral formation mechanisms (page 9194, lines 1-4). I think that the authors should extend their discussion on nucleation mechanisms. The intermediate ions formed via nucleation on ions and via attachment should have quite different properties in size distributions.

(1) While the clear gap in the ion size distributions (sub-class Ib.2) may indicate that neutral nucleation dominates at the measurement site, it could also be a result of the attachments of small ions to particles nucleated somewhere else (via either ion-induced or neutral nucleation mechanisms) and transported to the measurement sites. This should be pointed out in the first paragraph of page 9194.

(2) I would say that the obvious continuous ion distributions in the sizes range from ~ 1 nm to ~ 3-5 nm (sub-classes Ia and Ib.1, see Fig. 1) are clear indications of the dominance of ion-induced nucleation. This should be pointed out explicitly in the paper. Due to the rapid decrease of equilibrium charging fraction with decreasing size of nanometer particles, dominance of neutral nucleation should always lead to a clear

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gap in the sizes range from ˜1 nm to ˜ 3-5 nm.

(3) The authors have pointed out that sub-class Ib.2 days, which may be associated with neutral nucleation, are rare. Table 3 also shows that more than half of the BSMA identified particle formation event days belong to sub-classes Ia and Ib.1 which are clearly associated with ion-induced nucleation. Thus, it appears reasonable to conclude that at least half of the BSMA identified particle formation event days are clearly associated with ion-induced nucleation while only a few of the BSMA identified nucleation event days are probably associated with neutral nucleation. This should be pointed out in the text and reflected in the abstract.

2. In Table 2, the authors should add an additional column showing the number of days in each sub-class suggested by both BSMA (either BSMA+ or BSMA-) and DMSP measurements. Fig. 4 gives some information but it will be useful to list the numbers for different classes in the table for direct comparisons.

3. In Tables 2, 3, and 4, the authors presented number of days in each sub-class based on BSMA+ and BSMA- measurements. It will be very useful to add another column showing the number of days that both BSMA+ and BSMA- are in the same sub-class. This will give information about the number of days in each sub-class containing BSAM+ or BSMA- only.

4. After addressing comments 2 and 3 above, the authors may want to add some discussions on the possible reasons leading to the differences in the values derived based on BSMA+, BSMA-, and DMSP measurements. On page 9197 the authors gave several reasons briefly (including the suppression in the growth of nucleated particles associated with low concentrations of condensing vapors). The authors should give the concentrations of precursor gases measured to support the argument. Actually, it will make the paper more significant if the authors can present statistics on the meteorological (temperature, relative humidity, solar irradiance, etc.) and chemical (concentrations of SO2, H2SO4, NH3, certain organics, etc.) data measured during

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the same period. I believe that analysis of meteorological and chemical data will provide useful insight on possible reasons behind the difference in the values for each sub-class derived from BSMA+, BSMA-, and DMSP. For example, is there any statistical difference in key meteorological and chemical parameters between the days only BSMA- shows the event and the days only BSMA+ shows the events?

5. Page 9198, lines 12-14 and page 9200, lines 16-17. The authors emphasized that the negatively charged particle formation were generally more favorable at the measurement site. However, the data presented in the paper clearly show that nucleation on positive ions is also significant. I think that it is equally important to point this out in the text.

6. Page 9196, lines 4-6 and Page 9200, lines 1-3. The conclusion about the decrease in the relative number of DMPS detected particle formation events may change if the authors re-organize the year from April to March as suggested by referee #1.

Technical corrections:

Minor comments:

1. In the abstract, I would suggest that the percentage value (as shown in Tables 1-3) is also given in addition to absolute number of days.

2. Page 9189, line 22. With regard to the role of ions/charge on nucleation, the work of Yu and Turco (Geophys. Res. Lett., 27, 883-886, 2000) should be cited.

3. Page 9190, lines 8 and 10: change “to find” to “finding”.

4. Page 9191, line 7: change “summarised” to “summarized”.

5. Page 9193, line 16: “later shortly events” not clear.

6. Page 9197, line 9: “other” should be “are”?

7. Page 9197, lines 15-17. Since H2SO4 concentrations were measured, it will be

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useful if the authors can compare [H2SO4] for the growth suppressed cases with those in other cases.

8. Font in Table 1 is too small.

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