Atmos. Chem. Phys. Discuss., 5, S4993–S5001, 2005
www.atmos-chem-phys.org/acpd/5/S4993/
European Geosciences Union

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Conceptual study on nucleation burst evolution in the convective boundary layer – Part IV: Comparison with previous observations" *by* O. Hellmuth

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Received and published: 11 January 2006

1. General response to RC1-RC3

As for the evaluation of the previous parts, I would like to thank all three referees very much for the time they took to carefully scrutinise part IV.
For the arguments given in response to part I, the title will be changed as follows:
Columnar modelling of nucleation burst evolution in the convective boundary layer - First results from a feasibility study. Part IV: Valuation of the simulation results in view of previous observations

2. Summary of RC1-RC3

2.1. RC1

1. Page 11561, whole section: Whished to be commented on new nucleation parameterisation (in the light of the model results) given by Hyvoenen et al. (2005))

2. Page 11569, line 1: In Hyytiälä conditions it is found that concentration of H_2SO_4 is too low to explain the observed growth, see Boy et al. (2005). Since the growth from 1 to 3 nm is a crucial part in NPF, organics are very probable have at least some importance in NPF. This is in contrast with statements given in this article.

2.2. RC2

1. Page 11568, line 25: Ongoing research shows the importance of organics for NPF; Emission scenarios not yet understood; Statement of "no connection between photosynthetic activity and NPF" is too hypothetical

2.3. RC3

1. Page 11568, line 25: It has been shown by Boy et al. (2005), that H_2SO_4 can not explain the growth of the newly formed particle in Hyytiälä, and thus some other condensing agent, possibly organics, is needed. This would suggest an important part played by biogenics.

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3. Specific response to RC1-RC3

As RC1-RC3 are strongly correlated, I would like to answer in the common context. Among the open questions related to the nucleation issue, the role of organic chemistry in new particle formation, especially in forest regions, is one of the most complicated and trickiest. In the present paper, a number of empirical findings from the literature are cited, that support the binary case scenario. Among them were also some papers, that are related to observations of NPF in forests regions, especially based on Hyytiälä observations. As an argument seemingly supporting the binary case scenario, I cited the nice paper of Buzorius et al. (2001) (link between micrometeorology and NPF). Therein, the authors excluded a link between biogenic chemistry and NPF ...
Anyway, in their study the authors neither claimed, that this link is missing *always* nor missing *under all conditions*, observable or even typical in forests regions. The authors formulated *"no apparent connection"*, which is different from saying *"always no connection"* as insinuated in part IV. The "missing connection" is clearly related to the special conditions, under which NPF was observed in the study of Buzorius et al. (2001). The resulting misunderstanding in the present paper is awkward and deserves to be corrected. To my mind, there is no real contradiction between the findings of Buzorius et al. (2001) and that of Boy et al. (2005), Hyvoenen et al. (2005) and other authors on NPF in boreal forests.

All three referees agreed therein, that the corresponding paragraphs related to this issue should be revised, because some interpretations and conclusions seem to be in contradiction to recent findings on the role of organics in NPF in boreal forests (see details below).

In response to all three referees, the corresponding sentences were carefully revised (see details below). The papers of Boy et al. (2003c), Boy et al. (2005), Hyvoenen et al. (2005), as well as Lovejoy et al. (2004) were carefully evaluated and found to

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be so interestingly and instructive, that a more detailed evaluation is deserved. The questions adressed therein are related to a completely different mechanistic "way of life" for new particles. I have some ideas, how to setup both an "organic NPF scenario" and an "ion-mediated NPF scenario" using the present model approach.
However, I have realised, that my answer would clearly exceed the number of 15 pages allowed within ACPD interactive discussion. Therefore, I have decided to prepare a separate technical note to ACPD on that issue in connection with the interpretation of the present results. It deserves some refinements and will be submitted within the next few days. Hopefully, the referees can accept this way.

4. Changes of the manuscript

1. Page 11557, line 16:
   To be added:
   " ...(Eastern Germany). With respect to the boreal forest station Hyytiälä only such events were considered, for which no apparent connection between photosynthetic activity of the forest (a possible indicator of biogenic emissions of precursor gases for nucleation and condensation) and NPF was observed. For the considered events, one of the key hypothesis on the role of turbulence is confirmed."

2. Page 11558, line 20:
   To be added:
   " ... not unambiguous. With respect to NPF in boreal forests, the results must be very cautiously interpreted. The special conditions, under which NPF occurs there, especially the role of organic compounds are discussed. Nonetheless ..."

3. Page 11560, line 24:
   To be deleted:

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"Strong evidences for the binary NPF scenario"
To be rewritten:
"Certain evidences seemingly supporting the binary NPF scenario ..."

4. Page 11562, line 3:
To be added to the last sentence of subsection 2.1:
"However, the binary UCN evolution simulated here, must be cautiously interpreted with respect to observations of NPF in forest regions. As will be evaluated in more detail in a forthcoming paper, there are empirical findings from these regions suggesting an important contribution of organic compounds to both formation and growth of particles (Boy et al., 2003c; Boy et al., 2005; Hyvoenen et al., 2005). Unlike the positive correlation between water vapour and NPF in classical nucleation theory, in homogeneous nucleation of secondary organic aerosols, supposed to occur in boreal forests, water vapour and NPF are anticorrelated. This fact is related to the reaction of the stabilized Criegee intermediate with water vapour to produce high volatility compounds (Bonn et al., 2003; Berndt et al., 2003)."

5. Page 11568, line 25 - page 11569, line 3:
To be deleted:
"Importantly to note . . . and Buzorius et al. (2001, p. 399)."
To be rewritten:
"Evaluating CO_2 flux measurements Buzorius et al. (2001) concluded: *'Based on the CO_2 flux data, no apparent connection between the photosynthetic activity of the forest (a possible indicator of biogenic emissions of precursor gases for nucleation and condensation) and the particle formation occurrence was observed'*. As in the present model biogenic chemistry is neglected, special care has to be taken when assigning the model results to forest regions in general. Accompanying CO_2 flux measurements can provide valuable help to examine, whether the premises of the model with respect to biogenic processes are complied or not.

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Deviations from these conditions in boreal forest regions are discussed in Boy et al. (2003c), Boy et al. (2005), and Hyvoenen et al. (2005). There findings suggest an important role of biogenic compounds in NPF. Anyway, the simulated turbulent UCN flux pattern . . .."

6. Page 11577, line 7
   To be inserted between the preceding sentence ending with "... too." and befor the new sentence starting with "Altogether ..."
   "Particular cautiousness is advised with respect to the assignability of the simulated 'anorganic' scenarios to forest regions, where NPF and growth is apparently strongly correlated with biogenic emissions and organic compounds. An accompanying evaluation of CO_2 measurements can help to classify, which kind of scenarios must be considered."

5. Response to technical comments

5.1. RC1

1. RC1: Page 11569, line 15: "derive"
   AC: To be corrected in the final version. Thanks!

2. RC1: Page 11579, line 10: Year for citation Birmili et al., number 3 is missing.
   AC: Are you sure? I hope, we have the same text version. In my downloaded version the year 2003 is correctly cited on page 11579, line 12. The quotation of Birmili et al. (2003) on page 11560, 11570, 11571, 11572, 11573 is correct as well.

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5.2. RC2

1. RC2: Page 11559, line 24: "later on"
   AC: Will be corrected in the final version. Thanks

2. RC2: Page 11560, line 2: "photooxidation"
   AC: Will be corrected in the final version. Thanks

3. RC2: Page 11571, line 22: "later on"
   AC: Will be corrected in the final version. Thanks

4. RC2: Page 11572, line 14: "few orders of magnitude"
   AC: Intuitively, I would prefer "a few orders of magnitude" instead of using plural for magnitude. Let's decide a teacher. Thanks.

5. RC2: Page 11575, line 25: "to neglect"
   AC: Will be corrected in the final version. Thanks

5.3. Response to RC3

1. RC3: –
   AC: –

NEW: 3.3 NPF in forest regions

Using data from the Hyytiälä measurement station located in a boreal forest region, Boy et al. (2005) evaluated the contribution of H_2SO_4 to the particle diameter growth rates in the nucleation mode between 3 and 25 nm. For the growth rate

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fraction of sulphuric acid [1] an average value of 8.8 % was found (Boy et al., 2005, Table 5, empirically derived values ranging from 3.2 to 16.9 %). The authors concluded: *"Obviously sulfuric acid is involved (fraction between 3 to 17 % in new particle production and growth of aerosols over boreal forest regions in Northern Europe, although it might be not the key parameter in the particle formation process itself"* (Boy et al., 2005, p. 877). From previous long-term measurements at the Hyytiälä measurement site, Boy et al. (2003c) found, that $\mathrm{H_2O}$ vapour was anticorrelated with NPF. Water vapour is thought to affect NPF by two ways: (1) Enhancement of the $\mathrm{H_2O}$ vapour concentration will increase the real diameter of pre-existing aerosols, hence their volume and surface as well. Consequently, the condensation sink will increase, and the number of particles, measurable under dry conditions, remains limited. (2) As seen from laboratory experiments, the number of particles newly formed from the photolytic reaction of monoterpenes (α- and β-pinene) with ozone strongly decreases with increasing $\mathrm{H_2O}$ vapour concentration. More recently, the findings of Boy et al. (2003c) were confirmed by Hyvoenen et al. (2005). Form a long-term study using Hyytiälä measurements, they authors found, that the most important variables in explaining the nucleation events are the means of relative humidity (RH) and the logarithm of the condensation sink (CS). Relative humidity and NPF were observed to be anticorrelated. This effect can be mechanistically explained by the influence of organic chemistry.

Hyvoenen et al. (2005, p. 3354) concluded: *"Although we found a connection between the occurrence of nucleation and two key variables, the detailed chemistry still remenains speculative. One missing link in our study is the concentration of biogenic Volatile Organic Compounds (VOC) emissions, which are expected to be of high importance even at the low concentrations. . . . One possible cause of confusion is the possibility of two or even more different nucleation mechanism*

[1] That is the percentage of the growth rate, which is explained by sulphuric acid alone. The total growth rate is experimentally determined from measurements using a DMPS (Differential mobility particle sizer).

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acting simultaneously in the atmosphere. One such combination is clear-air nucleation vs. pollution nucleation, another possibility is combination of neutral and ion-induced nucleation." A detailed discussion of these findings with respect to the interpretation of the present "anorganic" scenario simulations will be given in a technical note prepared for Atmos. Chem. Phys. Discuss..

Interactive comment on Atmos. Chem. Phys. Discuss., 5, 11557, 2005.