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Interactive Comment

Interactive comment on “The Hohenpeissenberg aerosol formation experiment (HAFEX): a long-term study including size-resolved aerosol, H_2SO_4, OH, and monoterpenes measurements” *by* W. Birmili et al.

Anonymous Referee #2

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Review of the manuscript "The Hohenpeissenberg aerosol formation experiment (HAFEX): a long-term study including size-resolved aerosol ,H2SO4 ,OH , and monoterpenes measurements" by Wolfram Birmili, Harald Berresheim, Christian Plass-Dülmer, Thomas Elste, Stefan Gilge, Alfred Wiedensohler, and Ulrich Uhrner.

General comments: The manuscript is well written and disposed with clear figures. It is based on an important data set and includes a mature data analysis, important results and interesting conclusions. It should be accepted in Atmospheric Chemistry and Physics after some revisions. It was a pleasure to read the paper and I would like to apologize for being late with my review.

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Specific comments:

1. On page 1657, the authors list several nucleation and growth mechanisms recently proposed in literature, including (1) ternary nucleation, (2) growth activation of stable clusters, (3) ion-nucleation, and (4) nucleation by turbulent atmospheric mixing. A fifth possibility would be nucleation and growth induced by atmospheric waves (Nilsson et al., JGR 105, 2000), a process which unlike those suggested by Easter and Peters (1994) and Nilsson and Kulmala (1998) involves no turbulence and occurs solely in stably stratified air, but with a large potential to enhance nucleation and growth.

2. In section 3.2, the authors have used a clever way to estimate the horizontal extent of the aerosol formation. However, I believe that this method reveals the lower limit of the process, which with an average of 87 km and a maximum of 339 km places us on the mesoscale. In Nilsson et al. (Tellus 53B, 462-478, 2001) we demonstrated that the aerosol formation was observed simultaneous over three stations spread over 1000 km, which places the process on the regional scale and large parts of whole air masses.

3. It is a good intention by the authors to try to test if the nucleation and growth are similar at different vertical levels in section 3.2. However, with horizontal distance of 3 km between the instrument and a vertical difference of only 300 m, it is likely that the same air mass are either descending or ascending along the sloping terrain. The value of this comparison is therefore limited. The test is negative, no significant difference is found, and the authors conclude that there is a very homogeneous vertical distribution. Considering the terrain it is however more probable that the this shows that the size distribution and its evolution are similar over a horizontal distance of 3 km, than that it is similar over a distance of 300 m in the vertical. If there had been a difference, what would it have meant? The convective time scale in a mixed layer is 10 minutes (see e.g. Nilsson et al. in Tellus 53B, 441-461, 2001). If we consider horizontal wind speeds of 5-10 m/s, 3 km corresponds to only 300-600 seconds or 5-10 minutes time lag due to advection. This is similar to the typical convective time scale of the mixed layer and

to the observed time delay of 5 minutes. Therefore, it would both have been difficult to claim that a difference in the size distributions were due to vertical differences, when it could just as well have been due to advection, and it appears that the observed 5 minutes time lag may have been due to advection rather than vertical mixing. The authors should therefore be more careful with the conclusions they draw from this section.

4. Section 4.2, page 1665. I would like the authors to give more details on the normalisation of the solar irradiance in order to make sure that the same procedure can be repeated with other data sets for comparison in the future.

5. Section 4.2, page 1665. The observed relative humidity near surface is of very limited interest for the nucleation. On sunny days with convection present, air parcels will be cycled throughout the mixed layer and experience much high relative humidity at the mixed layer top, than near surface (e.g. Nilsson et al. in Tellus 53B, 441-461, 2001). The same applies to the temperature, but with opposite result (lower temperatures).

6. Section 4.2, page 1666. The result that the best nucleation events "in most cases" (please give a percentage) were "associated with the advection of southerly warm air masses that subsided in the vicinity of the Alpine mountain range" are important enough to be included in the abstract. Its also worth stretching in this section that this is opposite to the observations in norther Europe, where nucleation are associated with northerly cold Arctic and Polar air masses (Nilsson et al., Tellus 53B, 462-478, 2001).

7. Section 5.2, page 1668. There is actually a recent parameterisation available for ternary nucleation, easy to use and valid in a vide range of temperatures and vapour pressures: "Parameterisation of ternary nucleation rates for H2SO4-NH3-H2O vapors" by Napari, Noppel, Vehkamäki and Kulmala in JGR October 5, 2002. I suggest the authors expand section 5.2 to include also ternary nucleation. This would be valuable for the manuscript. The discussion in section 5.4 could then be shortened and included in section 5.2.

8. Section 5.3, page 1669. Why was the analysis restricted to the summer? I have

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difficulties to accept that "the boundary layer depth could be considered to be defined by thermal convection" only during the summer. The boundary layer is often even more convective during the spring, and it would have been very interesting to include the spring in the analysis since it contrasts to the summer as one of the seasons with much nucleation. Because the air in the spring is also colder, the adiabatic cooling in ascending air will reach lower temperatures and make it more likely for convection to trigger nucleation in the way suggested by Easter and Peters (1994) and Nilsson et al. (Tellus 53B, 441-461, 2001).

9. End of section 6.1 and part of section 6.2. It appears that those vapours that are most responsible for the growth are not the same as those most commonly responsible for nucleation. This conclusion could be emphasized. This result is very similar to the conclusions of the BIOFOR project, e.g. Janson et al. (Tellus 53B, 2001).

Any important changes in the manuscript caused by these comments, questions and suggestions should of course be reflected in "7 Conclusions" and perhaps also in the abstract.

Technical corrections: 1. Section 5.3, page 1669, line 18. "radio sond ascent" instead of "radioascent".

Interactive comment on Atmos. Chem. Phys. Discuss., 2, 1655, 2002.

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