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Atmospheric Chemistry and Physics Discussions

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 #1

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This paper reports 2 years of measurements of aerosol size distributions and gaseous concentrations of OH, H2SO4, and VOC (terpenes and aromatics) at a rural site in southern Germany. The purpose of the investigation was to investigate new particle formation and the role of the gaseous compounds in those events. The data are unique, data evaluation thorough, scientific questions of high interest to readers of ACP, and paper well written. The paper should be published. However, I do have a few specific comments I would like the authors to consider.

Specific comments:

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1) Section 2.4: It is stated that the measurement uncertainties for monoterpenes was 30-50%. Do you judge the results to be too high, too low, or just uncertain?

2) Section 3.1, line 9-10: It is stated that the concentration of ultrafine particles was generally low at night-time. Was there any NPF at night or was it exclusively a daytime phenomena?

3) Section 3.3 and Fig 3: It is stated that particle concentrations decreased at sizes below 5 nm and that the distribution was thereby "closed". You correctly point out the importance of ruling out measurement artifacts before trying to interpret this behavior. It seems that you do this but fail to take the next step. What is the explanation? Could the shape of the distribution curve at sizes below 11 nm indicate that the NPF is not occurring at the site, but rather some distance away, that the shape of the curve indicates transport and growth between times of nucleation and observation.

4) Section 4.2: It is stated and shown in Fig 7 that NPF anticorrelated to relative humidity. It would be of interest to test a correlation with absolute humidity.

5) Section 4.2: A short meteorological discussion is given in which it is stated that class I events during the winter were usually associated with warm air mass advection from the south. This is an interesting observation and should be elaborated on, especially in light of the findings at the boreal site in Finland where events were associated with cold air mass advection from the north (see Kulmala et al., 2001). Also, if I understand correctly, the mountain site is above the boundary layer/surface inversion during the night but within the boundary layer during the day, and that the inversion breaks up around 1000 h. Could the mixing which occurs during the breakup, i.e. the mixing between the nocturnal boundary layer air and the residual layer air above, be important for NPF? Is there any correlation between the time for observation of NPF and the time for the breakup of the nocturnal boundary layer?

6) Section 5.4: It is stated that, by ternary nucleation, (H2SO4/H2O/NH3), "high nucleation rates would be predicted from measured H2SO4 on most days", that this "is not

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in agreement with the observations", that "scavenging intensity by pre-existing particle" might be the reason for the relatively low frequency of NPF events, and finally that "no uniform picture evolved from the HAFEX data." However, Fig 7 shows that the median for CS is greater than 4x10-3 during the warm season and lower than this during the cold season. Can you identify a critical CS or particle surface area, above which no events occur? Might this explain the lack of events when concentrations would otherwise suggest NPF by ternary nucleation? It seems you have the data to investigate this question further. Secondly, could the fast growth rate during summer (section 6.1) make it more difficult to observe NPF, see comment 7.

7) Section 6.1, 6.2, 6.3 and Fig 14, 15, and 16: It is shown in section 6.1 and Fig 14 that that observed growth rates were much higher in the summertime than in the wintertime, and the conclusion is drawn that "this result is in contrast to the reduced frequency and intensity of new particle formation in summer." You need to distinguish between nucleation and growth. Might the larger growth rate be a reason for the lack of summertime observations? If we interpret the closed distribution shown in Fig 3 to indicate that nucleation occurs not locally, or at least not primarily locally, but at a distance from the site, then couldn't a result of the faster summertime growth rates be that UF particles are not often observed. Nucleation may still be going on, but the new particles will have grown/coagulated to beyond the UF size class before you get to measure them. So instead of the increased summertime growth rates being in contrast to reduced intensity and frequency of NPF, it may explain the reduced observation of NPF in summer.

8) Section 7, Conclusions: Line 15, p 1675: "the calculated terpene turn-over rates did not show a significant correlation with the missing particle growth rate". This was not shown or discussed in the article. On the contrary, in the last paragraph of section 6.3 it is stated that "the HAFEX results confirm a clear seasonal link between the production rates of biogenic aerosol precursors and the growth rates of newly formed aerosol particles." This point needs to be cleared up.

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Interactive Comment

Technical comments

1) Section 4.2, line 13: subsidised should be subsided

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