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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.

W. Birmili et al.

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Author response to anonymous referee # 1

1. *"Section 2.4", "measurement uncertainties for monoterpenes""*
   The monoterpene concentrations are considered to be uncertain within +/- 30-50 % of the values indicated. Text has been added accordingly.

2. *"Section 3.1", NPF an exclusive day-time phenomenon?*
   Over the entire observation period we never oberved any NPF event at night.

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We therefore conclude that solar radiation is required to produce burst of new particles at the Hohenpeissenberg site, and that any possible night-time sources are negligible in comparison with this day-time production. This point has been clarified in the text.

3. *"Section 3.3", "closed" size distributions*
   Since there has been considerable interest in this discussion, we decided to add a new section (Section 7) to the manuscript, providing discussion on the possible implications of the particle size distribution shape. We considered this discussion useful near the end of the manuscript because it builds upon the experiences made in Sections 3–6. The end of Section 3.3 was also changed accordingly.

4. *"Section 4.2", absolute vs. relative humidity?*
   Besides those of relative humidity, diurnal profiles of absolute humidity were now calculated according to the reviewer's suggestion (see "Fig. 8" of the manuscript). As a result of this analysis, a significant text passage was also added to section 4.2:

   "Profiles of absolute humidity suggest that in the warm season it was largely a low absolute humidity which caused the low RH and particularly, not excessive temperatures. In the cold season, there was a also significant contribution of warm temperatures (warm air advection) leading to the low RHs observed during NPF events of classes I and II. Since humidity is no independent factor (it correlates with cloudiness), the physical interpretation of this factor in relation to particle formation is not clear."

5. *"Section 4.2"*
   Aspect A: "warm air masses from the south"
   We have thought about the apparent mistery of NPF in air masses from the south, as opposed to air masses from north during BIOFOR. Our conclusion is that these two observations do not necessesarily represent a contradiction if, for in-

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stance, northerly air implies clean air during BIOFOR (with low pre-existing surface area, and instable stratification), and southerly air implies similar conditions during HAFEX.

A particular difficulty during HAFEX is the interpretation of air movements and exchange at the Hohenpeissenberg site, only several ten km north of the Alpine mountain range. The observed events of warm air advection, for example, point to Foehn conditions, or Foehn-like conditions which, in turn, may be associated with subsidence of air in wave packets, or stress-induced mixing by breaking lee waves. These phenomena are, again, very difficult to judge without the availability of three-dimensional data. Further, back trajectories may lack accuracy in the vicinity of such inhomogeneous terrain. As the result of an earlier examination of the mentioned aspects, we prefer to keep our relatively general statement about the southerly air masses.

Nevertheless, the text at the end of Section 4.2 was modified to clarify this point:

"Further evidence indicated the advection of south-westerly or southerly warm air masses during these events, originating mostly from Southern France and the Mediterranean (class I: 5 out of 5; class II: 15 out of 19). Being ca. 30 km north to the Alpine mountain range, the Hohenpeissenberg site is then prone to be influenced by Foehn or Foehn-like *conditions, which can lead to the subsidence of air in wave packets, or stress-induced mixing by breaking lee waves. These phenomena, which would be favourable for the formation of new particles were, however, difficult to assess using the limited spatial information available to this study. The back trajectories used probably also lack accuracy in this strongly inhomogeneous terrain and prevent confident localisation of the source regions of these air masses. We can therefore not decide if the low particle surface area alone, or the mentioned orography-related phenomena were responsible for the observation of the associated NPF events.*

Aspect B: "mixing during the break-up of the boundary layer"

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It is a promising idea to search for correlations between the occurrence of new particles and indicators of vertical air exchange. Especially in the cold season, when the mixed layer is shallow, one would expect the observed particle formation to take place the closest to the measurement site as possible throughout the year (cf. also the discussion forwarded in the companion paper, Uhrner *et al.*, 2002, ACPD). To verify this aspect, we used the concentrations of nitrogen oxide (NO) as an indicator for the break-up of the boundary layer. NO originates mainly from the anthropogenically polluted layer of air below the Hohenpeissenberg mountain. Once the developing mixed layer starts wrapping the Hohenpeissenberg mountain, a significant increase from near zero-levels of NO to a maximum of typically 0.5 ppbV. Due to the less intense mixing, higher midday concentrations are reached in the cold season. To compare the onset of new particle formation and the time when the mixing starts, the increase of both the UFP and NO curves were parametrised (UFP: exponential function; NO: Gaussian function), and the times of the half-peak-height were compared. The results, however, show that there is no strict correlation between the observation of NPF and the suggested time for the break-up. Data points close to the curve of unity suggest that the two processes coincide. The data is, however, too scattered to allow a solid conclusion. Unfortunately, the evolution of the vertical mixing process at the site cannot be characterised with the exactness required for such a rigid temporal comparison.

6. *"Section 5.4", "Could you identify a critical CS or particle surface area (...) ?"*
We had checked this aspect, however, with no conclusive result except for the cold season. To clarify this point, a new Figure ("Fig. 8") was added to the manuscript showing the statistical distributions of CS for the various event and seasonal cases. It can now be clearly seen that a considerably lower CS is associated with the NPF events in winter, compared to the non-events in winter. This aspect has been slightly expanded within the text, and the aspect of reduced CS

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in southerly air masses is now also included in the abstract.

7. *"Section 6.1, 6.2, 6.3", "Observed growth rates"*
You are right suspecting that higher growth rates could lead nucleation mode particles to grow out beyond the size range $3 < D_p < 11$ nm, which is our definition of "ultrafine particles". Model calculations showed that coagulation becomes a rather slow process once the particles are larger 8–10 nm, with particle lifetimes tending towards several hours against coagulation. Condensation would therefore be the remaining dominant process altering such a nucleation mode. It does, in contrast to coagulation, not diminish the particle number concentration. If a nucleation mode grew beyond the size of say 11 nm, it should therefore still be visible by a concentration peak in the size spectrum at sizes large than 11 nm.

   We also observed particle formation events where the main particle burst appears at relatively large particle sizes, with the maximum particle concentration occurring at > 11 nm from the beginning of the event. We have anecdotal evidence that this type of NPF events occur mainly in the warm season, which would be consistent with the increased capacity, e.g., of monoterpenes to cause nucleation mode growth. Over the 2.5 years, however, we only observed a "handful" of these cases, so we deemed a separate event class not to be justified (Nevertheless, they are an interesting object of study!). Regarding the detection of these events: Concentrations still exceed > 1000 cm^{-3} in the interval 3–11 nm, so they are being recognised by the filter using UFP concentrations, and classified as an event of class I or II. In conclusion, the possibility that a nucleation mode grows extremely fast in the summertime, and thus escapes detection is unlikely.

   In relation to this issue, the research group from Lund has recently reported interesting observations of particle size distributions in the Amazon rainforest (Zhou *et al.*, 2002, *J. Geophys. Res.*). Measurements of NPF events there on the ground showed an even more emphasized trend towards large sizes in the nucleation mode diameter. When new particles were formed in the Amazon rainforest, they

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tended not to be seen in the size distributions before having reached a size of ca. 15 nm. This observation would plausibly correspond to a scenario where particle growth occurs relatively fast (presumably due to abundant organic precursors) so that the measurements miss a nucleation mode below < 10 nm, but they detect it at larger sizes.

8. *"Section 8", "...correlation with the turn-over rate"*
The text was indeed formulated ambiguously. Basically, two differing results were produced in this paper with respect to this issue: 1) There is a *seasonal* correlation between the particle growth rate and the monoterpene turn-over rate. This correlation needs to be referred to in climatological terms and does, in particular, not refer to individual days. 2) Meanwhile, there is *no* significant correlation between the two when one looks at a series of individual days. By analysing the latter aspect, we had hoped to find such a correlation, which would have provided a satisfactory explanation for the involvement of the oxidation products of organics in nucleation mode growth. These points were accordingly clarified in the Section 6 (new subsections introduced) and the Conclusions.

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