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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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Authors' response to anonymous referee 2

1. *"On page 1657...", nucleation and growth mechanisms*"

   In accordance with the comment,we have completed the list of mechanisms by that of atmospheric waves, including the citation Nilsson et al. (2000).

2. *"In section 3.2.", extension of NPF events*

   It is true that this method probably yields a lower estimate of the air masses'

dimension in which NPF takes place. The manuscript text was accordingly modified:

"This yielded an average extension of 87 km (minimum: 6 km, maximum: 339 km) suggesting that the NPF events in the central European source region extend over the mesoscale. Since at a fixed measurement site the full extent of these air masses might not be captured, we consider the range of a few 100 km as a lower estimate.

On the other hand, I found no indications that 1000 km range observed in Northern Scandinavia can be generalised, for instance, for Central Europe. Previously, I compared two sets of concurrent particle size distribution measurements with respect to the simultaneity of NPF observations. One set considered measurements over 6 months (spring and summer) at the sites Leipzig (urban) and Melpitz (rural) in Eastern Germany. (The Leipzig data was provided by B. Wehner, personal communication). The two sites were 50 km apart, in flat terrain. The coincidence of NPF events at both sites was expectedly high, above 90 %. The second set of observations covered observations over 2.5 years at Hohenpeissenberg (this data), and Leipzig (urban), distant at ca. 500 km in north-south direction. A coincidence of NPF events was, in this case, hardly existent and not different from accidental coincidence. It must be noted that the sites 500 km apart were also separated by several chains of medium-level mountains, representing an effective barrier for the weather. I think that in the presence of such orography, and also in the noticable presence of concentrated anthropogenic plumes, the zones where NPF happens may probably be smaller than "air masses" after their classical definition as synoptic-scale structures in temperature and humidity.

3. *"It is a good intention...", multi-level observations*

   Your arguments are convincing. The statement has therefore been weakend from referring to a vertical into general spatial variation:

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"In a attempt to characterise the spatial homogeneity of NPF events, particle size distributions were measured concurrently at MOHp (980 m) and a second site at the foot of the Hohenpeissenberg mountain (680 m). Fig. 2 shows the evolution of a NPF event, manifested by two separate waves just after 1030 h and 1200 h, which were detected simultaneously at both sites. The horizontal wind speed was 5 ± 1 *m s*$^{-1}$*, blowing perpendicularly to the line connecting the two sites. The particle size distributions and total particle concentrations evolved in a very similar fashion at both sites (separated 3 km horizontally and 300 m vertically), demonstrating a relatively uniform spatial distribution of the NPF event in the inhomogeneous terrain around Hohenpeissenberg.*

4. *"Section 4.2, page 1665", normalisation of solar irradiance*

   The text in the manuscript has been improved in order to better illustrate this aspect:

   "In order to make a seasonally invariant distinction between cloudy days and clear skies, each daily solar irradiance cycle was normalised by a cloudless reference profile. Reference profiles were obtained by averaging the 3 profiles of a particular month of the year that showed the most amount of radiation. A radiation value of 1 accordingly refers to a clear sky."

5. *"Section 4.2, page 1665", relative humidity*

   We consider the thermodynamic situation at the ground of some interest since we still have no final experimental evidence where exactly the new particle are formed. An increased humidity and decreased temperature at the mixed layer top might be offset by a corresponding dilution of precursor gases, although we do not know very much about the latter aspect. LIDAR profiling in the continental boundary layer (Wandinger *et al.*, 2002, *Journal of Geophys. Res.*; A. Ansmann, personal communication) suggests that there is always a significant vertical gradient in (optically visible) aerosol concentration, despite even distributions of water

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vapour indicating perfect mixing. This could also apply to gaseous constituents with variable sources on the ground. There is certainly a strong need to investigate this issue with improved profiling methods, and from airborne platforms.

The comment is also addressed in the rewritten Section 5, where the ternary nucleation rates are compared between ground level and mixed layer top, and the new Section 7 ("Discussion of the closed particle size distributions") that emerged from both, your and referee # 1's comments.

6. *"Section 4.2, page 1666", advection of southerly air masses"*

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 instance, northerly air implies clean air during BIOFOR (with low pre-existing surface area, and instable stratification), and southerly air implies similar conditions during HAFEX. To improve on this aspect, the text in Section 4.2 has been rewritten as follows:

"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 (Georgelin et al., 1997) 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."

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Unfortunately, the meteorological data available to us (synoptic scale data) is too rudimentary to warrant further conclusions, and also a sound comparison to BIOFOR. We therefore prefer to keep the relatively general statement about the southerly air masses. Nevertheless, the observation is now included in the abstract.

7. *"Section 5.2, page 1668", ternary nucleation*

   The parametrisation of ternary nucleation, not yet published at the time of writing, has now been included in all calculations instead of binary nucleation. Accordingly, the Sections 5.1–5.3 have been completely rewritten, the figures been replaced, and the conclusions been modified. As suggested, section 5.4 became obsolete. Using ternary nucleation, however, the atmospheric ammonia concentration arises as an unknown parameter. Based on empirical observations of ammonia at another site in Germany, and on the upper limit of Napari et al.'s parametrisation, we selected an ammonia mixing ratio 100 pptV for the scenario at Hohenpeissenberg (see also motivation in the text).

8. *"section 5.3, page 1669", boundary layer convection*

   Indeed some data were lacking in the Figure, and there has been a misunderstanding as well. In our analysis, "warm season" spans the months between March and October, so it includes the "spring" when the boundary layer is particularly active. To make the analysis more sound a criterion involving the mixing layer has now been included:

   "Importantly, only cases were considered when the mixed layer height was greater than 700 m (44 out of 64; overwhelmingly cases from the warm season, i.e., the months March-October). This height corresponds to 1.5 times the vertical distance between the Hohenpeissenberg mountain and the radio sonde measurement site (in flat terrain), and ensures that the MOHp site was within the mixed layer."

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As a result, the Figures 12–13 have been altered accordingly.

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