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Interactive comment on "The contribution of sulphuric acid to atmospheric particle formation and growth: a comparison between boundary layers in Northern and Central Europe" *by* V. Fiedler et al.

Anonymous Referee #2

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Interactive comment on "The contribution of sulphuric acid to atmospheric particle formation and growth: a comparison between boundary layers in Northern and Central Europe" by Fiedler et al

This article focus on typical quantities associated with nucleation events at two different measurement sites located in Hyytiälä (Northern Europe) and Heidelberg (Central Europe). Measurement campaigns have been performed at the two locations during the same period (spring) for two subsequent years. Observations of aerosol number size

distribution, sulphuric acid concentration as well as meteorological parameters are presented. In addition, by using trajectory analysis a description of typical flow conditions prevailing during nucleation days is supplied in the manuscript. The authors are using established methods to calculate particle growth rates, formation rates and source of condensable vapours and compare the results between the stations. The contribution to aerosol growth from sulphuric acid is calculated and the results are compared between the stations. The article is well written and proper English is used throughout the manuscript. The issues addressed and the results presented in the article are clearly within the scope of ACP and the article merits publication after some minor changes are included.

General comment:

In the article, the authors evaluate and compare some key-features associated with nucleation events at two different stations. One of the stations represents clean background conditions and the other one represent highly anthropogenic influenced conditions. The calculated parameters require the assumption that the air-mass observed is homogenous on relatively large scales during the observations (i.e. permits an eulerian approach to the problems). This is probably a good approximation for background stations such as Hyytiälä that is affected by a minimum of local sources. However, some questions are raised by the fact that an evidently large number of local point sources and population densities surround the MPI station. It is therefore some concern regarding the possibility of comparisons of the calculated quantities between the two stations. This does not only concern the particle formation rate but also the growth rates as well as other calculated parameters relying on homogenous air-mass properties. Some of this concern would clearly be shattered by more detailed description of the size distributions evolution at the two sites by for example adding some “classical” surface plots describing the size distribution evolution at the two stations and the diurnal variation at large as well as more detailed description of the origin of the air during the nucleation events.

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Specific comments:

On page 574 lines 13-15: “The gaseous sulphuric acid lifetime with respect to condensation on aerosol particles ranged from 2 to 33 min in Hyytiälä and from 28 s to 8 min in Heidelberg.” It could be wise to mention the concept of lifetime and its derivation from CS also somewhere in the manuscript and not just the abstract.

Page 575 line 4-26 the authors say that number size distribution from 3-900nm was observed at both stations. On the link supplied on page 576 line 13 I am informed that the Hyytiälä size distribution only covers 3-500nm. Does this discrepancy between size ranges of the instruments somehow affect the calculated parameters? E.g. if the DMPS systems cover different size ranges at the different stations, is the CS integrated over the same size interval at both stations? Does it matter at all?

Page 578 line 18: “To reduce statistical errorsĚ” Although I understand the meaning of this line I found it initially hard to follow. Please rephrase.

On page 579, section 2.3.3 the authors describe how particle formation rate is calculated. I can agree with the method, but it is not accurate to refer to these calculations as nucleation rate as is done on page 583 line 2. The method described does not give the rate of nucleation, but rather the rate of change of particles 3-25nm. In order to calculate actual nucleation rate the whole spectrum of aerosol dynamic processes leading to the appearance of 3-25nm sized particles has to be considered.

Page 579 line 19 the authors state that the calculation of particle formation rate not is valid when different air parcels are transported to the station. What about local point sources? I think the suggested approach is more useful at background locations such as Hyytiälä. As pointed out in some places in the manuscript the location of the MPI measurement site makes the measurements likely to be influenced by local sources. Is it under these circumstances fruitful to calculate particle formation rates at all? Combustion processes and vehicles are strong emitters in the 3-25nm size range.

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On page 580 starting on line 19 the authors state that a strong decline in particle concentration could be observed during most nucleation events. However, in following paragraph (Page 581 lines 3-6) the authors say that boundary layer evolution during morning hours usually brought polluted air to the Heidelberg station as the boundary layer enveloped the station. This is somewhat contradicting. Did nucleation days lack this feature? Please clarify.

Page 583 line 3-7: The authors say that the highest formation rates are observed during class 1 event days which in a way seems reasonable. However, on line 5-7 they conclude that this is a result of low pre-existing aerosol concentrations, leaving more vapours participate in nucleation. Following this statement one would expect class 1 nucleation days to be associated with lowest CS. Looking at table 1 there is no clear relation between nucleation class and CS.

On page 583 lines 26-29 the authors say that percentage contribution from sulphuric acid to particle growth rate 1 (GR1) always seems to be the same, independent of location. Does a comparison of a limited number of particle formation events between two single stations really support this statement? Especially since calculations of growth rates at MPI Heidelberg may be strongly biased by local emissions.

Interactive comment on Atmos. Chem. Phys. Discuss., 5, 573, 2005.