**Atmospheric
Chemistry
and Physics
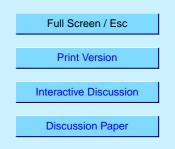Discussions**

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Comment

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 #1

Received and published: 14 March 2005

This paper by Fiedler et al. addresses an important unsolved topic, the mechanisms of the initial steps of formation and growth of atmospheric nanoparticles. More specifically, in this case the role of sulphuric acid is investigated, using data from field experiments conducted at two sites, Heidelberg (Germany) and Hyytiälä (Finland).

The paper is written in good english, is clear and for the most part, also logical and scientifically sound. However, the obtained experimental data still need some further chew-up to support the conclusions stated.

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Major comments:

1. I don't understand the role of figures 4 and 5 in the paper, which show the amount of radiation and temperature as functions of time. These results are not used anywhere in the paper and, if not somehow tied to the other results or conclusions presented, add no value to the paper. As such, figures 4 and 5 should be removed.

2. One of the main conclusions of the paper is that the existence at Hyytiälä and non-existence at Heidelberg of the correlation between the concentrations of the smallest particles (3nm-6nm) and sulphuric acid. This is, however, shown (figures 8 and 9) only for one single day at Hyytiälä and one at Heidelberg (where there actually is a clear correlation). This is not enough. Since the scatter plots of N3 vs. H2SO4 have been obviously plotted for each day, why not calculate a correlation coefficient for each day and add it to the result tables. Then, any reader can believe the stated conclusions.

3. On page 586, it is stated that air mass trajectories were analysed and some fuzzy conclusions given based on these analyses. Again, why not show these results? It would not take much effort/space to add a column to the result tables showing the air mass direction. This would make the already written discussion about the trajectories much more convincing.

Minor comments:

4. On pages 578-579, two methods for growth rate estimation are given. For the first method, it is unclear if the actual nucleation mode is fitted with a lognormal distribution, or are the temporal evolutions of the individual size class concentrations fitted? Please explain more clearly! Is this method self-invented or can you give a reference?

5. In the second growth rate estimation method on page 579, the nucleation size is specified as 1 nm. Why? Can this approximation cause uncertainty in the analysis?

6. When estimating particle formation rates on page 579, it is assumed that the concentration of particles between 3 nm and 25 nm is affected only by the apparent nucleation

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into this range. What about scavenging? or growth outside of this range? It is possible that scavenging can have a significant effect especially at Heidelberg. Please quantify these effects somehow.

7. In many places in the text, the term nucleation is used falsely, when actually is meant apparent nucleation of particle formation to the above 3 nm range (e.g. in table 1). Please change term and use e.g. J3 instead of J as the symbol.

8. At the beginning of page 580, a relationship is given between Cvap and GR. This must involve an assumption of the vapor or vapor properties. Please specify. Can this assumption cause large errors in the further analysis?

9. In equation 2, a maximum flux is assumed, e.g. the saturation vapor pressure of the condensing vapor is assumed zero. In some of Kulmala's earlier but recent work the nonzeroness of this term is used to explain observed size-dependent growth rates. Thus, is equation 2 in its presented form applicable?

10. What is the uncertainty in the sulphuric acid measurements?

11. At bottom of page 581, it is stated that 'For our purposes, the diurnal variation of total radiation was taken as a surrogate for UV-B radiation.' What are these purposes? The presented data are used nowhere.

12. At bottom of page 583 (and in conclusions) the authors state that sulphuric acid contributes the same percentage to particle growth, independent from region. This is by far too strong of a statement and should be replaced by stating that it contributes the same percentage at both places (not all possible places in the world)!

13. Figures 8 and 9 should be in similar form. Why is there a shifted curve in fig. 8 but not in fig. 9 ? How is the time-shift chosen? Also, it wood be a good addition to show one 'bad' day, especially from Heidelberg, since it is stated that for many days in Heidelberg there was no correlation. Furthermore, the correlation coefficients for the days in the figures should be given.

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14. In the conclusions, on page 586, the difference between Hyytiälä and Heidelberg regarding size dependent growth rates in discussed. The authors say 'Both may be explained by the influence of other gaseous substances...' This is confusing and needs further clarification.

15. In the abstract, the authors give lifetimes, but all the analysis in the text is in terms of sinks. Why? Please use same terminology in all parts, or, explain the relationship between these two variables.

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

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