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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Atmospheric particle formation events at Värriö measurement station in Finnish Lapland 1998–2002" *by* H. Vehkamäki et al.

H. Vehkamäki et al.

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Response to referee's comments:

Referee #1: Scientific questions/issues

The authors make extensive use of event classification, quantification of formation rates (cm-3 s-1), and quantification of growth rates. The numerical conventions used by the Finnish group should be documented to encourage uniformity in these types of calculations by other groups. The methods (if already published) should be cited, or if unpublished, should be included as an Appendix. This is not required, but I think would be useful to the nucleation measurement community.

Reply: Wehave added references to the text "We estimated the formation rates of 8 nm particles and their growth rates from the size distribution data using the methods presented by Mäkelä et al. 2000 and Kulmala et al 2001."

Minor points 1. In the abstract, the sentence beginning (The air masses) is confusing.

Reply: The sentence has been changed to "The air masses, with clearly elevated sulphur dioxide concentrations (above 1.6 ppb) came, as expected, from the direction of Nikel and Monschegorsk smelteries."

2. In the green oval in Figure 3, the numbers 147 and 135 seem to be transposed.

Reply: yes,there was a mistake, this has been corrected

Referee #2:

Specific comments:

1. The detection limit and measurement uncertainty of at least the SO2 monitor should be given. This is because of the relatively low SO2 concentrations frequently measured at the remote site, as well as because the degree of pollution is classified according to S1471 SO2 concentrations (section 2, last paragraph).

Reply: The detection limit has been added to the text which now reads: "The measurements for trace gases (sulphur dioxide SO2, ozone O3, nitrogen oxides NOx), temperature, absolute humidity and wind speed were also performed continuously at 2.2 m, 4.4 m (not trace gases), 6.6 m, 9 m and 15 m levels of the measurement tower adjacent to the cabin. SO2 was measured with a fluorescence analyser (Model 43S, Thermo Environmental Instruments, Inc., detection limit 0.1 ppb, maximum error +/- 0.05 ppb). A detailed description of the other trace gas measurements ..."

2. On page 3541 (lines 5-12) it has been speculated that one reason for the night time events might be the sulfuric acid originating from aged pollution in which practically all SO2 has already converted into sulfuric acid. In the atmospheric boundary layer, the time scale for condensation of sulfuric acid from gas phase to pre-existing particles is always considerably shorter than the time scale over which SO2 is oxidized into gaseous sulfuric acid. As a result, sulfuric acid concentrations cannot be very high if SO2 concentrations are low (regardless of the pre-existing aerosol concentra-

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tion or condensation sink). The only way we would reach high gaseous sulfuric acid concentrations is SO2 depleted air would be evaporation of sulfuric acid from the particles. This is possible but not very likely. The authors should correct this point in their manuscript.

Reply: the text has been changed to "One possibility is that these trajectories brought aged polluted air from North America or Britain, and the particle formation was caused by an unknown vapour which could not be detected with the current instruments in Värriö."

3. "2 m or 2.2 m height". What does this mean? Are there two height separated vertically by 20 cm?

Reply: Trace gases, temperature and wind speed are measurend at 2.2 m, relative humidity at 2m next to the DMPS system. The sentence has been corrected to: "For trace gases and temperature we used the value measured at 2.2 m and for relative humidity value at 2 m height, which was close to the particle measurement level."

4. It is stated that "higher water vapour concentrations in the air seemed to prevent particle formation" (page 3542, line 12). The authors should be more careful about the cause-effect relation. There is clearly an association (anti-correlation) between these water vapour concentrations and particles formation events, yet I doubt it can be said that the high water vapour concentrations prevent particle formation.

Reply: The sentence has been changed to: "High water vapour concentration in the air seemed to anticorrelate with particle formation, as has been ..."

5. In "Conclusions" it is stated that "most of the event occurred during the spring months between March and May" (page 3544, lines ,16-17). This is not true when looking at Figure 1. The number of events between March and May was approximately 70 which is less than a half of the total number of events (147).

Reply: The sentence both in abtstract and conclusions has been changed to read:

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The frequency of particle formation and growth events was highest during the spring months between March and May

6. A comparison to other studies concerning the frequency of observed events, their seasonal variation and the associated particle formation and growth rates should be made beyond the extremely short comparison performed in section 3 (second paragraph; pages 3538-3539). At the very least, the comparison should cover observations made at approximately similar latitudes in Northern Europe (mainly Finland and Northern Scandinavia) and North America.

Reply:Text has been extended in two places:

Chapter 3: "The frequency of the formation events exhibits a clear spring maximum with just over 0.2 events per day in May, as can been seen from Figure 1. The spring maximum is also typical for other Boreal sites like the SMEAR II station in Hyytiälä, southern Finland (Mäkelä et al., 1997). However, there it occurs already in April with around 0.4 events per day, consistent with the fact that spring starts earlier in a more southern location. The weaker autumn maximum in the particle formation event frequency that is observed in Hyytiälä (0.15 events per day in September) can be seen in Värriö in August (above 0.1 events per day), but it is not as clearly distinguishable. Throughout summer and autumn the number of events per day In Värriö is below 0.1, decreasing to only less than 0.03 events per day during the winter months. In Hyytiälä the event frequencies are somewhat higher, close to 0.1 in the summer and around 0.05 events per day during winter Urban St. Louis (Shi, 2003) exhibits an more scattered picture with a minimum around 0.05 events per day during midwinter, and the highest values above 0.3 events per day during April, July and September. Opposite to the other sites Hohenpeissenberg in (Birmili et al., 2003) rural central Europe exhibits the maximum with over 0.25 events occurring during midwinter, and minimum (around 0.075 events per day), during late summer and early autumn. It seems that very different processes are controlling the particle formation events in different locations there are clear, but the are clear similarities between the two boreal sites Hyytiälä and Värriö."

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Interactive Comment

Chapter 6: "We estimated the formation rates of 8 nm particles and their growth rates from the size distribution data using the methods presented by Mäkelä et al. 2000 and Kulmala et al 2001. The particle growth rates varied between 0.5 and 10 nm/h (Figure 9a). The growth rates have a maximum in summertime and minimum in wintertime. The formation and growth rates in December, January and February are left out since there are only a few data points for these months. Also in Hyytiälä, Hohenpeissenberg and St. Louis the growth rates have a summer maximum and winter minimum, and event their values are very similar (Kulmala et al., 2004). The formation rates were around 0.1 cm-3s-1 (Figure 9b), and they exhibited no clear seasonal variation. These formation rates are lower than those typical in Hyytiälä (0.5-1 cm-3s-1), and more anthropogenically affected measurement sites like Hohenpeissenberg and St. Louis. Furthermore, the growth rates and the formation rates did not seem to correlate with each other. This finding supports the conclusion of several other studies (see Kulmala et al., 2004). "

Figure 1: events per day has been added for easier compsrison with figures in Kulmala et al., 2004

7. Finally, although the manuscript is relatively well written, there are still a number of grammatical errors that should be corrected before publication. Reply: The text has been corrected by a native English speaker.

In addition to the changes listed here we have made some minor corrections to the text and figures.

Interactive comment on Atmos. Chem. Phys. Discuss., 4, 3535, 2004.