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Interactive comment on “VOC measurements within a boreal forest during spring 2005: the role of monoterpenes and sulphuric acid in selected intense nucleation events” by G. Eerdekens et al.

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General comments

The manuscript by G. Eerdekens et al. investigates two distinctive aerosol “nucleation events” at the well-known Hyytiääla field station. For this purpose, they present an extensive set of measurements during both events and from these observations they derive different mechanisms to be responsible for particle formation in each case. Given the still many open questions on different possible mechanisms of aerosol formation, this paper is certainly of interest for ACP and a welcome contribution. My main general criticism is that the paper presents too many data in rather great detail, that are later

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discussed as being not so relevant for the focus here, which makes the manuscript too long and hard to read. On the other hand, important data for supporting some of the conclusions, namely the investigation of enantiomers of monoterpenes, is only mentioned as part of the discussion. It is therefore recommended that the authors streamline their manuscript and maybe place part of the data presented here as supplementary material in order to make the manuscript shorter and more concise. Apart from that the results are presented carefully and correctly interpreted, with two exceptions that are addressed in the specific comments below: 1) I was not convinced by the given information why “event 2” is truly an event of particle nucleation, 2) The support of monoterpene enantiomers for identifying the saw mill as major source needs to be extended.

The authors finish their conclusions suggesting saw mills to be of substantial importance for air chemistry and secondary aerosol formation in boreal forests. A short discussion on the frequency and spatial scale of such saw mill influences should be added to support such a strong statement.

The manuscript is suitable for publication in ACP after the specific comments have been addressed by appropriate revisions.

Specific comments

General comment to graphs: They tend to be overloaded and insufficiently labeled which makes it difficult to grab the essential information quickly.

Fig.2: Improve labeling of different time series for better distinction (e.g. press vs. WANG ?, wind direction?). Height of Tdew measurement? Gap in RH data?

Fig. 3: Generally too small, difficult to see diel variations and nocturnal (evening) peaks of monoterpenes. How was the 10 min. average done when measurement of one profile took 20 min?

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P12792, I 15f: The discussion about methanol/acetone could be omitted as it does not add to the main focus of the paper and as acetonitrile was low, an influence of biomass burning seems unlikely.

P 12794: I am not too familiar with the classification scheme for particle formation events, but the “event 2” seems no obvious nucleation event. Both figures 6 and 12 suggest that there was no development from smaller to larger particle sizes during this event, which I thought was a mandatory criterion for showing evidence of new particle formation. What is the reason why event 2 was classified as nucleation event and not simply as a situation of sequential advection of air masses heavily loaded with particles of all sizes interrupted with short moments of cleaner air?

P. 12794: Paragraph 5.2. There is not sufficient information on how the box and whisker plots were generated. Supposedly they were computed by grouping all night and day time concentrations of the respective height. In this case it is not surprising that any information on potential gradients is lost as day to day variation in concentrations are likely to be much higher than any vertical differences and the considered period is not sufficiently long that these day to day variations would be filtered out. A more meaningful way may be to look at normalized profiles.

Fig. 7: Show acetone/methanol plot as discontinuous line

P. 12795, I 27: The change in methanol/acetone on 17:00 is not sharp and clear in comparison to the variation of this ratio throughout the rest of the day.

P. 12796: Methane results should be placed in the discussion of Fig.8. (or be omitted completely)

P. 12796: Discussion of wind direction/speed: Figure 7a shows wind directions at 3 heights but these are not discussed. On the other hand, wind speeds at different heights are discussed in the text but no data are shown.

P. 12796, line 25f: It makes more sense to plot RH instead of absolute humidity. For

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the latter it is sufficient to note it did not correlate with particles (without showing).

P. 12797 | 12f: The interpretation that polluted air was advected cannot be followed from the given information on ozone concentrations. How different is ozone from polluted and unpolluted air at this site?

P 12797 and Fig 9. The link of SO₂ and the event is not obvious to me, neither is the significance of the city of Jyväskylä as an SO₂ source in this particular event. Maybe it is just the simplicity of the "plain fetch" that is irritating me. It makes the reader attempt to find the SO₂/sulph.acid signature of the city in the concentrations, which cannot be found. As this discussion is not central for the main topic of this manuscript, I suggest simply stating that this city may be the SO₂ source in the case of continuous transport of air masses from that ENE sector.

Figure 10: The gap in meteorological parameters just prior to the first particle burst is unfortunate (reasons?), but all lines should be drawn discontinuously in such a case, as it is misleading, especially since only a part of the corresponding parameters were drawn in this way.

Discussion of Event 2 As in the case of event 1, RH is discussed in the text, but the figure only shows absolute humidity, please change.

P.12799, last paragraph: As noted before, in this event there is no visible development of smaller to larger particles, hence no nucleation event.

P. 12801, line 27. The term "footprint" should be avoided in this context. The respective discussion for event 1 looked at species with longer lifetimes, which will always exhibit a much larger "footprint" than short-lived species. Similar to my comments to Figure 12, the argumentation should be simplified in order to avoid misunderstandings. The different wind directions and signatures in VOCs and other (longer-lived) compounds speak for themselves and provide enough evidence for the hypothesis of the saw mill as an important factor in event 2, but not in event 1.

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P 12802, line 20: See comment to vertical VOC profiles above.

While many of the above discussed parameters and potential roles in particle formation have been described previously, the investigation of enantiomeric monoterpenes represents a novelty at this site. The presentation of these results as part of the discussion seems somewhat awkward. Also, the support of the enantiomeric ratios for suggesting the saw mill as the source of the monoterpenes in event 2 is not clear from this discussion. There are at least 3 other periods in Figure 13 with the same or more pronounced enantiomeric signatures of the monoterpenes as in event 2. These events should then also be easily connected with advection of air masses from the saw mill (or others?). If not, these previous extended periods with +/- ratio of 2.5 suggest that there are other possibilities to cause strong signals in the enantiomeric composition.

I am missing a discussion on how frequent and how widespread these “locally caused” events are, before stating that “sawmill industry substantially can influence air chemistry and formation of secondary aerosol particle over the boreal forest”.

Technical corrections

P. 12792, l 12: Events should also be marked on figure 3.

Fig. 4: What do colored areas/error bars represent (std. dev.? quartiles?)

Fig.5: Add significance of solid and dashed lines also in figure caption.

P 12794: l 8: replace "and" by "until"

P. 12796, l 22: Obviously something is missing here.

Fig. 7: Show acetone/methanol plot as discontinuous line

Figs 7 and 8: Add a title in the legend of the profile measurements indicating the respective parameter. Only mentioning it in the caption is misleading.

Fig 8b) Indicate the height of the measurement of the sum of monoterpenes.

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