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Atmospheric Chemistry and Physics Discussions

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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We are very grateful for the astute comments and suggestions of the reviewer. We believe we have addressed all of the issues raised in the revised version.

The detailed responses to the reviewer's comments are given below.

The paper is now entitled

"VOC measurements within a boreal forest during spring 2005: on the occurrence of elevated monoterpene concentrations during night time intense particle concentration events."

General Comments

As suggested by the reviewer, the manuscript has been shortened and slightly reorganised. We have improved all graphs for readability and labelling in order to avoid further confusion. There are no unreferred graphs in the paper, and apologise for the previous oversight.

As stated above we have amended the title to avoid confusion on this point. The title has been changed to "VOC measurements within a boreal forest during spring 2005: on the occurrence of elevated monoterpene concentrations during night time intense particle concentration events."

Specific comments

Title

1. Corrected as stated above.

Introduction

2. We agree with Riipinen et al., (2007) that clear nucleation events are those events which are clearly detectable through particle number and growth. The disputable sentences have been rephrased and the text currently reads as follows: "Several aerosol nucleation events have been analysed by Riipinen et al., (2007) for this campaign and the authors concluded that the main process limiting the observable particle formation was the competition between the growth of the freshly formed particles and their loss by scavenging, rather than the initial particle production by nucleation of sulphuric acid. Other particle formation events, in which sulphuric acid and water vapour are not key components, occurred synchronously with rapid and unexpectedly strong increases in monoterpenes"

Experimental

3. The stated accuracy is 2% absolute of the actual reading.

Results

4. We now refer to studies by Sellegri et al., 2005 and Taipale et al., 2008 who have reported diel cycles for spring time characterised by elevated nocturnal VOC mixing ratios. However, neither have reported monoterpenes mixing ratios which have been as high as reported here.

5. Figures 5 and 6 have now been assigned according to their occurrence.

6. We refer to Fig 7b which now includes the wind speed as it is important in the transport of particles. The superfluous labels P, Q, R, S have been removed.

7. DMPS spectra between April 24 and April 29 show 2 modes in the aerosol size distribution: a fine and a coarse fraction (similar to Figure 9a). The fine mode can be related to the new particle formation events and is highly variable. The coarse mode also indicates a shift to larger particles over the abovementioned period and which potentially relates these particles to the nucleation events, the particle concentrations in this size range is less variable. Coarse mode data were averagely distributed between $\sim$20 nm and $\sim$200 nm (not 110 nm, typo in the text).

8. We agree with the reviewer and the sentence has been removed.

9. The reviewer is correct to note the inconsistency in the description of Figure 2. The lowest aerosol particle concentrations (400 to 500 cm-3) appeared in the dataset around the onset of the nocturnal boundary layer for the period between April 18 and April 23. Afterwards, total particle number concentration has been consistently higher than 1000 cm-3 until May 1. Rather than describing the background particle concentration we have found it more appropriate to discuss the condensational sink which is a measure for the time, which a sulphuric acid molecule has until it collides with the next aerosol surface area.

10. The paragraph has been shortened and now reads ”Daytime and night time vertical VOC gradients were not substantially different, except for the monoterpenes.”

11. As reviewer 2 correctly remarks, there have been nucleation events preceding the night time events on April 25, 26 and 27 which have not been mentioned at all as they are being discussed in by Riipinen (2007) in great detail. There also has been a similar, weak nucleation event on April 24. This nucleation event has not been followed by night time bursts in the particle number concentration. All four events have each been all characterised by a shift in the particle size distribution towards larger particle diameters over the course of time (typical banana shape in the DMPS-spectra). From Figure 5 it can be seen that there has been overlap of these events. The elevated level of particles lasted until the breakup of the nocturnal boundary layer on April 29. Particles produced during these daytime nucleation events could very well have acted as nucleii for organics to condense on and thereby grow to bigger sizes. However, as the particle number concentration during Event 2 (only referring to the intense peaks) has been much higher (about 9 times) than observed for the nucleation event. Due to the stratification of the air it is very well possible that isolated air parcels with origin over the saw mill have been advected over the measurement station.

12. The ammonia measurements for this campaign have been described and discussed in Riipinen 2007. Unfortunately, ammonia measurement were stopped on April 23 and therefore not discussed in detail in this study.

13. We now describe more precisely that H2SO4 was found to be anti-correlated with the particle concentration between the start of Event 1 and the strongest increase in the particle concentration around midday. Squared correlation coefficients now indicate how well certain tracers were correlated with each other.

14. Wind speed and wind direction discussed here as well as the mentioned angular deviation reflect the situation above the canopy (so above 16 m). There have not been measurements of the wind speed and direction at 4.2 m. Data from the 8.4m have not been considered as they are largely influenced by the forest (too much friction) making it very hard to interpret or link to the origin of the airmass.

15. A correlation between m69 and m93 was only found during the night time events under discussion. Although there has not been additional proof from the TD-GCMS measurements, we strongly believe that the increases in m/z 69 and m71 during Event 2 were very unlikely related to natural emission of isoprene and its oxidation products given the time of the emissions and squared correlation coefficients for m/z 71 and m/z 69 with toluene (m/z 93) were 0.61 and 0.59, respectively. Therefore it seems more likely that isobaric compounds or fragments of compounds at m/z 87 have caused the increase in the signal at m/z 69. Our measurements indicate that there has been a stronger contribution the second and third peak of Event 2 onto m/z 69 than to m/z 87 compared to the first peak in the series.

16. As shown above the title of the paper has been changed so as to avoid confusion regarding nucleation events. We have amended the text to indicate that we describe variation of monoterpenes during intense particle concentration events rather than completely elucidate their formation as the evidence we have in indirect.

Technical Corrections

1. Both amended as suggested.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 12781, 2009.