

Interactive comment on “Understanding the formation of biogenic secondary organic aerosol from α -pinene in smog chamber studies: role of organic peroxy radicals” by B. Bonn et al.

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Received and published: 31 May 2007

Comments to reviewer #1

General comments

We need to thank reviewer #1 for his very valuable comments concerning our manuscript and we will include his suggestions throughout the study in order to improve the quality.

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1. The reference of the EU-OSOA project final report [3] is certainly a non-reviewed reference and difficult to access. However, to our knowledge the investigated experiments have not been included in a previous publication. Because of that we aimed to refer to all participating scientists within the OSOA project. In the revised version a detailed description of the measurement of ozone, α -pinene and NO_x will be given and an intercomparison of measured and simulated data will be included. This will certainly improve the readers understanding and clarify the statements given.
2. The second point refers partly to the first one and tackles the possibility of ozone particle interactions, i.e. reactions. It is true that ozone was present in excess during the entire experiment, thus at the time of the second injection too. Nevertheless, α -pinene has a single double bond only. Ozone is generally not assumed to react with any other bond of the terpene than a carbon double bond [2]. Since the terpene won't partition significantly to the aerosol phase because of its high vapour pressure (ca. 5 hPa), the double bond will oxidize in the gas-phase before entering the aerosol phase. A possibility for ozone to interfere with the aerosol might only be the reaction with nitrate compounds. However, the nitrogen oxides (NO and NO_2) concentration was fairly low and thus the concentration of nitrates within the aerosol too. Because of this the effect of this kind of interaction was neglected, which will be stated in the revised version.
3. α -pinene measurements and simulated concentrations will be given in a new Figure in order to allow a direct intercomparison and thus to give more background information.
4. With respect to the water content, the aerosol was assumed to be dry. This

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relies on the measured relative humidity of -42°C (0.015%), which is much below the deliquescence point. This will be named as well as the discussion of the labile/stable fraction as a function of particle size extended. Thanks to the reviewer for pointing this out, because this it among the most crucial results obtained and it should be understood by the reader. From Figure 4 it is already shown that the labile fraction is most likely highest in the smallest size sections and decreasing towards larger sizes due to (a) increased partitioning of 'stable' compounds with increasing particle size and (b) because of the oligomerization process, transferring labile compounds into less 'fragile' and more 'stable' oligomers.

Minor comments:

- Table 2: The first formula includes two organic peroxy radicals, i.e. C1079O2 and C96O2, because of the reduced chemical scheme. They are not formed in the very first reaction step, which forms the primary ozonide, but in one of the subsequent steps. However, since the first reaction of the terpene with ozone is the time limiting step, these reaction steps have been reduced to a single one [1]. The notation is kept close to the Master Chemical Mechanism to guide the user [4]. C96O2 is the organic peroxy radical formed by the OH reaction with α -pinene and C1079O2 is the sum of C107O2 and C109O2 produced within the hydroperoxide channel [2]. We will add a comment to explain this in more detail.
- Correct. The axis legends have been lost during the transfer between different file types and the modification of the boundary box. This will be taken care off in the revised version.

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- As stated in the text the injection of α -pinene causes a higher concentration on the local scale before the injected terpene is sufficiently diluted within the entire chamber. During this stage particle formation already occurred. These processes are difficult to tackle with the box model. Therefore we assumed in the initial stage a higher α -pinene mixing ratio to allow nucleation to occur. However, the dilution in the entire chamber causes the number concentration not to reach the one corresponding to 11.4 ppbv but to 9 ppbv. This is not a linear process, which resulted in an approximately six times smaller number concentration. This needs to be explained more clearly in the text and it will be done in the revised version.
- OK. The expression 'mass diameter' is a shortening of 'mass derived diameter'. Assuming a spherical cluster of a density of $1200 \frac{\text{kg}}{\text{m}^3}$ this results in a diameter of 0.95 nm. This will be written in the extended version.

References

- [1] Bonn, B., von Kuhlmann, R., and Lawrence, M. G.: Influence of biogenic secondary organic aerosol formation approaches on atmospheric chemistry, *J. Atmos. Chem.*, 51, 235–270, 2005.
- [2] Finlayson-Pitts, B. and Pitts, J. N.: *Chemistry of the upper and lower atmosphere*, Second Edition, Academic Press, New York, 2000.
- [3] Hoffmann, T. (ed.): *Origin and formation of secondary organic aerosol*, Final report, EU project, Bruxelles, 2002.
- [4] Master Chemical Mechanism (MCM) v3.1: <http://mcm.leeds.ac.uk/MCM/>, 2007.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 3901, 2007.

ACPD

7, S2173–S2177, 2007

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