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Interactive comment on "Nanoparticle formation by ozonolysis of inducible plant volatiles" *by* J. Joutsensaari et al.

J. Joutsensaari et al.

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We thank Prof. Claeys for her review and believe that the paper will be strengthened by our revisions prompted by the comments.

1) Page 2 (abstract) - line 13. The authors write: "Our results therefore suggest that atmospheric nucleation events proceed via condensation of oxidized organics on preexisting molecular clusters rather than via their homogeneous or ion-induced nucleation." This statement is too strong in my opinion: the results obtained in the present study rather suggest that atmospheric nucleation events proceed through a mechanism that is different from homogeneous or ion-induced nucleation of oxidized organics. It cannot be claimed that results have been obtained suggesting that this mechanism involves

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"condensation" of oxidized organics.

- Our statement of course is based on the assumption that oxidized organics are involved in atmospheric nucleation events in one way or another, i.e. that they participate either in the nucleation or growth, or both, of the particles. It is generally believed that oxidized organics are indeed involved in the nucleation events. However, the evidence for this is indirect, e.g. the behavior of 3 nm particles in condensation nucleus counters indicates that their composition is partly organic (O'Dowd, CD, P. Aalto, K. Hämeri, M. Kulmala, T. Hoffmann: Aerosol formation: Atmospheric particles from organic vapours, Nature 416, 497-498, 2002). Insofar as there is no direct evidence, the participation of the oxidized organics in atmospheric nucleation remains a hypothesis. We will explain this in the revised manuscript.

2) Page 3 (introduction) - line 1. Instead of citing the review article by Kanakidou etal., 2004 here, it would be more appropriate to cite an original research article; for example, the study by Kavouras et al. [1], who provided the first field evidence for secondary organic aerosol formation from alpha-pinene, could be cited here.

- We will cite Kavouras.

3) Page 3 (introduction) - line 17. The comment made above under 1) also applies here. I suggest to weaken this statement.

- See above.

4) Page 4 (experimental) - line 3. Here, I would write: " initiate an emission spectrum of terpenes that is typical to "

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- OK

5) Page 4 (plant material and treatments) - lines 13-14. For the sake of the interested reader, who may not be familiar with the chemical nomenclature of terpenes, I suggest to give some explanation between parentheses here. For example: monoterpenes (C10 terpenes) homoterpenes (acyclic terpenes derived from sesqui- or diterpenes) sesquiterpenes (C15 terpenes)

- OK

6) Page 5 (aerosol formation experiments) - line 16. Filtered pressurized air is used in the chamber: can it be excluded that this air does not contain SO_2, which may oxidize and as such provide sulfuric acid molecular clusters on which new particle formation may occur? This is relevant to what is written later in the paper where it is stated that no sulfate clusters are present in the plant chamber experiments. Was SO_2 measured in the experiment?

- We did not measure SO_2. Even if some SO_2 had managed to get through the filtration of the pressurized air system, OH-radicals would be needed for the oxidation. Since no UV light was used, any OH radicals would be "dark", i.e. formed as a byproduct of the ozonolysis reactions, and the OH levels created in such a way certainly cannot be very high. Taken together, it is extremely difficult to believe that sulfuric acid would have formed in any significant quantities.

7) Page 7 (plant experiments) - line 5; Table 1. Looking at the results presented in this Table, I wonder whether the data presented for 3-hexen-1-ol + 2-hexenal and 3-hexenyl acetate for cv. Rinda have not been interchanged; it is not logical that the induced levels are lower than the controls.

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That is a right conclusion. We have had this odd result with the Rinda variety in our earlier experiments also. The listed compounds are the “C6 green leaf volatiles” that are released just after the mechanical damage to plant tissues. The leaves of this variety are very sensitive for handling. When the plants were fitted in the VOC collection cuvettes some small fractures appeared and induced emission of these compounds. Interestingly, Rinda plants treated with MeJa were not any more so sensitive to emit these compounds. We have observed this same sensitivity of intact control plants to handling and rapid release of C6 compounds with some broadbean (*Vicia faba)* varieties in herbivore experiments. We will shortly discuss this observation in the revised version.

8) Page 7 (plant experiments) - line 15. The authors write: “Globally the main monoterpenes are alpha-pinene, beta-pinene and limonene ”. Again, it would be appropriate to cite an original research article; I suggest to cite Guenther et al. [2].

- OK

9) Page 7 (plant experiments) - line 16. This is a suggestion for future experiments (no action to be taken for the manuscript): the authors argue that the cabbage emission spectrum of terpenes is more relevant to atmospheric conditions than the VOCs or mixtures of VOCs commonly used in smog chamber experiments. I suggest that for future experiments they consider a system that also includes isoprene, which has emissions on a global scale that are higher than those of the monoterpenes and which accordingto recent field and laboratory studies also serves as a precursor for secondary aerosol formation [3-8]. Furthermore, it has also been demonstrated that there is a co-variance between isoprene emissions and new particle formation above a coniferous forest [9].

-This is a good suggestion. Poplars, Norway spruce and *Sphagnum* moss are all iso-

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prene emitters and we currently monitor volatile emissions of these plant species. In fact, we have observed that hybrid Poplar (*Populus tremula x tremuloides*) saplings damaged by moth larvae have isoprene emission on the level which is about 10 times higher than the total monoterpene emission observed from MeJA-treated plants in this study.

10) Page 9 (aerosol formation experiments) - line 18. Here, I would write: " we believe our experimental results are relevant to atmospheric conditions." (instead of "are in fact consistent with")

- OK

11) Page 9 (aerosol formation experiments) - line 8. The comment made above under 1) also applies here. I suggest to weaken this statement.

- See above.

12) Page 10 (conclusions) - line 23. As already pointed out above, to better assess aerosol forming capacity of vegetation, it is essential to evaluate the whole emission spectrum of different plant species, not only including sesquiterpenes and other inducible terpenes, but also isoprene. As argued under comment 9) there is sufficient field and laboratory evidence that isoprene participates in secondary aerosol formation.

- OK

13) Page 10 (conclusions) - line 14. The comment made above under 1) also applies here. I suggest to weaken this statement. The results suggest that homogeneous nucleation that is observed in the plant chamber experiments does not operate in particle

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formation in the ambient atmosphere. It remains to be demonstrated that the latter mechanism simply involves "condensation" of oxidized organics on pre-existing molecular clusters.

- The term "condensation" is admittedly too specific, the growth of the particles could e.g. result from absorption/adsorption plus subsequent chemical reactions of organic species. We will clarify this in the revised paper.

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