

# **Deliquescence, efflorescence, and phase miscibility of mixed particles of ammonium sulfate and isoprene derived secondary organic material**

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## **General:**

The manuscript describes influence of isoprene photo-oxidation products on deliquescence and efflorescence of ammonium sulphate particles, observed in a laboratory study. In contrast to monoterpene oxidation products isoprene oxidation products have a significant effects on DRH and ERH. This can be important in the atmosphere as the mixed particle will contain water over a larger range of RH compared to ammonium sulphate / monoterpene-SOA particles or pure ammonium sulphate. The experimental procedures applied are sophisticated and as a consequence the analysis and interpretation is complex. The main task was to entangle the different organic loadings distributed over the seed particles. Although the authors clearly take efforts to transport the information in ordered way, the paper was in parts very difficult to read. I attribute that to the large degree of formalization in the description e.g. the logical table in the supplement. I would suggest a more descriptive way. However, the results derived are excellent and the conclusions are sound. Overall the paper is acceptable though and it should be published in ACP after the authors have addressed a few minor issues.

## **General comments:**

1.) Complex procedure info is often mixed with extra info in brackets is distracting. It would be helpful first to get a straight description of procedures, then details and less important modifications of the previous statements. This regards especially the first paragraph of section 2.2.1 and line 8 – 23 on page 9918.

The logical table provided me more difficulties in understanding than it helped. Especially, since the authors switched between RH and epsilon as describing parameters in the final statement in the manuscript.

I also had difficulties with the notation “scanning of epsilon” which was never scanned, instead a band width of epsilons contributed to the observations (page 9918, lines 8 or 20).

2.) I think the authors should comment/exclude the following issues in the manuscript regarding potential artifacts:

Are there new particles formed in HEC in absence of ammonium sulphate and isoprene when H<sub>2</sub>O<sub>2</sub> is photolyzed? If yes, how much mass is formed and is this of organic nature ?

Is there SOM formed in presence of ammonium sulphate and in absence isoprene when H<sub>2</sub>O<sub>2</sub> is photolyzed ? If yes, how much mass is condensing on the sulphate seeds and is this of organic nature ?

Are there new particles formed when isoprene is oxidized in the presence of ammonium sulphate?

3.) The authors mainly compare the inflow of sulphate particles to the outflow of mixed particles in the manuscript and in Fig. S1. I think, it is necessary to show the comparison of sulphate particles in the outflow of the chamber in absence and presence of isoprene oxidation products.

## Minor remarks / errors:

Page 9904, line 16 : Definition of  $\xi$  ?

Page 9909, line 1 : Why was the SMPS operated at 25% RH?

Page 9911, line 8 : At several instances the authors talk of stepwise changes of  $d_{\text{mono}}$  and report two diameters. Were these two diameters selected or were the diameters stepwise changed between these limits? See also page 9912, line 26 or Table S1.

Page 9911, line 21 – page 9912, line 8 :

This is an interesting consideration, but it should go into supplement. The experiments were defacto performed at diff.conditions.

Page 9912, line : Isn't that a "classical" hygroscopic growth experiment? Why do you call number-diameter distribution experiment ?

Page 9915, line 12ff : Here a sketch (in the supplement) would support faster understanding.

Page 9916, line 6 : I don't understand this statement. "Exchanging dependent and independent variables, the line  $\text{DRH}(\epsilon)$  appears in Fig. 4."

Page 9917, line 13 : "Extrapolation of the DRH line of Fig. 4 to <15% RH suggests a crossover between cases *b* and *c* at an organic volume fraction close to 1.0." I don't see why Fig 4 is suggesting this.

Page 9920, line 19 : I think it should read "Fig. 3c".

Page 9922, line 25ff : On oxidation isoprene likely forms small molecules which are highly oxidized compared to the large mono- and sesquiterpenes with less oxidized products. Isoprene products with O/C 0.4 may not even stay in the SOM.

Can it be that isoprene oxidation products have stronger effects on ERH than the small molecules applied in Bertram et al. 2011? If so, what do you suggest as a reason? It may well be that C/S and O/C ratio are not sufficient to classify deliquescence and efflorescence behaviour, instead the specific functionalization is of importance.

Page 9923, line 12 : I doubt it. Are there indications that Aikens parametrization of retrieval of O/C ratios does not apply to isoprene oxidation products. Your results suggest that O/C of the isoprene oxidation products is larger than determined by AMS.

Page 9934, line 24 : page numbers of reference Smith et al. 2011 are not correct.

Page 9938, Fig.2 :

Violet point is not at  $\epsilon = 0.6$ .

Why do use here the notation "positive" and "negative perturbation". Why not using "increased" and "decreased RH".

Page 9940, Fig.3, Caption, line 9 : replace "red lines" by "red dashed line"

Suppl. Table S1: "For simplicity, adjacent RH values that are equal are condensed to a single RH value in the table." I don't get the point here.