

Interactive comment on “In-cloud processes of methacrolein under simulated conditions – Part 3: Hygroscopic and volatility properties of the formed Secondary Organic Aerosol” by V. Michaud et al.

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We thank both reviewers for their careful examination of our work and for providing us with recent and interesting literature references which, we hope, will definitely help us ameliorate the quality of this paper.

We now will try to answer to the reviewer suggestions one by one:

"This paper is part 3 of a larger body of work, however if treated as a solo entity not enough information is provided as to why methacrolein SOA is important; a few sen-

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tences are placed in the summary but more should be written in the introduction." We now added a few sentences in the introduction which better address the importance of studying methacrolein.

"This work is unique in that it studies specifically methacrolein aqueous photo-oxidation SOA formation but the results are similar to and can be correlated with other SOA ageing studies. For instance, Perri et al, 2009 observe the production of low volatility organic acids and oligomers via aqueous photo-oxidation. Meyer et al, 2009 and Asa-Awuku et al, 2009 also observed that more volatile compounds appear to be more hygroscopic. Meyer et al, 2009 found VH-TDMA measurements of unseeded SOA show a decrease in the hygroscopic growth (HGF) factor for increased volatilisation temperatures. Asa-Awuku et al, 2009 quantified the effect of changes in volatility and hygroscopicity for the CCN behaviour of beta-caryophyllene SOA. Incorporating these and other findings (see specific comments) will strengthen this work's importance and implications" These references will be added in the text.

COMMENTS "1.What is the temperature of the water in the nebulizer? Is it the same as the aqueous solution used to generate the SOA? If not what kind of an effect will it have on the volatility of the atomized systems?" The water in the nebulizer was at the room temperature (constant at 20°C) while the aqueous solution in which SOA was generated was maintained at 25°C. It was shown in Liu et al, this issue (part 1) that when the light is off, no more reaction takes place in the reaction tank. Hence, the change of temperature between 25 to 20 °C does not imply additional chemical reactions. The temperatures are so close that we do not expect any impact on the volatility of the atomized particles. This information is now included in the text.

"2.What is the ambient temperature? And how does it range within the experiments? How can we compare values in Table 2 if Tambient is significantly different?" The ambient temperature is kept at 20°C.

"3.How does adding salt to the aqueous solution after the reaction validate that the

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SOA is not influenced by the presence of inorganic salt? If the SOA contain polar compounds, interactions with the positive and negative ions could be of importance once salt is added to the solution (e.g. ion-dipole interactions, salting out effects)." No chemical effect due to the presence of NaCl is detected in our measurements, as witnessed by the closure between the measured and calculated hygroscopicity of the NaCl+SOA mixture. This is now specified in the text.

"4. The work of Warren et al, 2009 found SOA (from gaseous-phase oxidation) was altered in the presence of gaseous water during their hygroscopicity measurements. In addition, Volkamer et al also found SOA formation in the aqueous phase was not limited to cloud droplets, but proceeded in the absence of clouds. How much would additional ageing during H-TDMA and V-HTDMA affect the composition of methacrolein SOA material in this study? The authors refer to properties characterized in Part 1 (Liu et al) and Part 2 (El Haddad et al), Can it be assumed that we are characterizing the same material? Any assumptions should be clearly stated in the text." Some chemical reactions due to the presence of water vapour may occur during the residence time of the SOA in the humidity controlled part of the VHTDMA. However, these reactions would occur during less than a few seconds in the dark, which should be put in parallel to an preceding in-cloud reaction time of several hours with irradiation. Our assumption is of course that the reactions which would take place in the VHTDMA are negligible regarding the reactions which we are studying over several hours in the liquid phase. It would not have been the same if we had studied dry condition SOA formation.. We are now specifying this point in the text.

"5. The authors state, " the aerosol physical properties change due to aqueous phase photooxidation has never, to our knowledge, directly been quantified with identified compounds". The measurements of Sorooshian et al, should be quoted. For example, Sorooshian et al, 2007a found "aqueous phase reactions to produce organic acids, mainly oxalic acid, followed by droplet evaporation is a source of elevated organic acid aerosol levels above cloud. Oxalic acid is observed to be produced more efficiently

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relative to sulfates the cloud liquid water content increases, corresponding to larger and less acidic droplets." Sorooshian et al, 2007b also found similar results. We meant by "physical properties" the aerosol hygroscopic and volatility properties, and we are now more specific in the text." Although Sorooshian et al. did not characterize these properties we found their work very useful and we are now referring to it.

SPECIFIC COMMENTS: "Please be consistent with the word aging and ageing." Done

"P6453 L7: The direct and indirect effects are both dependant on. . . The statement reads as though size distribution and hygroscopic properties are the only properties. Include some of the factors mentioned in following sentences (e.g composition, optical properties)" Done

"P6453 L14: Are the authors referring to only known mixtures of organics? It is unclear. If not there are several SOA ageing experiments that belong here and that are mentioned later on (e.g Varutbangkul, 2006)" We clarified this sentence

"P6453 L22: Replace "the resulting ambient" with "the resulting aged ambient"" Done

"P6453 L28: Previous VHTDMA technique and measurement papers should be cited as well (e.g. Johnson et al, 2005)." Done

"P6454 L8: Along the same lines as Kalberer et al, 2004, Perri et al, 2009 can be cited here." Yes we cited them in the next sentence and in the conclusion as well.

"P6454 L17 and L21: "However the aerosol physical properties . . . " Include the works of Sorooshian et al." As already mentioned, we meant hygroscopic and volatility measurements of SOA. Sorooshian et al. did not perform such measurements, so they can not be cited here.

"P6454 L19. Chen et al have also shown that methacrolein is an important precursor for SOA and Fu et al, 2009 have demonstrated the importance of the aqueous phase reactive uptake for SOA formation. This is related to the statement that methacrolein can form SOA and will reinforce its significance for this study." We now mention that

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Chen et al. 2008 already found in-cloud formation of SOA from methacrolein oxidation.

"P6460 L5: replace "Jin" with "In"" OK

"P6460 L8: replace "equivalent" with "similar". 1.8 seconds and 2 seconds" Done

"P6460 L10. Replace " we evidence" with "we see evidence"" Done

"P6463 L2: Methacrolein is misspelled." Done

"P6467 L5: Include chen et al reference" Chen et al. 2008 performed caloratory experiments while we meant natural cloud droplets. We now precise this

"P6467 L5: Replace "which seem to be different" with differ done

eTable 5: Please add a Table annotation for the abbreviation ND. Figure 1: Text in figures is very small , blurry, and difficult to read." Done

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 6451, 2009.